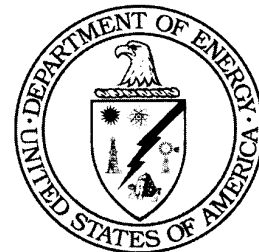


ENVIRONMENTAL MANAGEMENT  
& ENRICHMENT FACILITIES  
MANAGEMENT AND INTEGRATION  
CONTRACT

**RECYCLED URANIUM  
MASS BALANCE PROJECT  
PADUCAH GASEOUS  
DIFFUSION PLANT  
SITE REPORT**

**June 14, 2000**

**Final Draft**



MANAGED BY  
BECHTEL JACOBS COMPANY LLC  
FOR THE UNITED STATES  
DEPARTMENT OF ENERGY

**RECYCLED URANIUM MASS BALANCE PROJECT**  
**PADUCAH GASEOUS DIFFUSION PLANT**  
**SITE REPORT**

**DRAFT FOR REVIEW**

Prepared for the  
U.S. Department of Energy  
Office of Environmental Management

BECHTEL JACOBS COMPANY LLC  
Managing the  
Environmental Management Activities at the  
Paducah Gaseous Diffusion Plant  
Under contract DE-AC05-98OR22700  
For the U.S. Department of Energy

# RECYCLED URANIUM MASS BALANCE PROJECT

## PADUCAH GASEOUS DIFFUSION PLANT

### SITE REPORT

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# **RECYCLED URANIUM MASS BALANCE PROJECT PADUCAH GASEOUS DIFFUSION PLANT SITE REPORT**

## **Executive Summary**

During various periods of Paducah Gaseous Diffusion Plant's (PGDP) operating history, uranium (U) recycled from the Department of Energy (DOE) reactors used to produce plutonium and tritium has been introduced to the PGDP operating cycle. The recycled uranium (RU) primarily originated from the DOE reactors at Hanford and Savannah River. Intermediate processing of some of the Hanford and Savannah River U also occurred at the FMPC site and Oak Ridge Gaseous Diffusion Plant (ORGDG) prior to shipment to PGDP for further processing. A lesser quantity of RU introduced to PGDP originated with foreign sources. The primary concern with the RU is that it introduced plutonium (Pu) and neptunium (Np) (transuranic (TRU)) products and technetium ( $^{99}\text{Tc}$ ) (a fission product (FP)) to the U processed at PGDP and increased the potential hazards for both workers and the environment.

This Mass Balance Report has been prepared as a part of a DOE complex-wide initiative for the review the mass flow characteristics of the RU over the past 50 years. The authority for this initiative is documented in Deputy Secretary of Energy, T. J. Glauther's memorandum of September 15, 1999. This memorandum documents the following project goals applicable to the mass balance activity:

- Identify the mass flow of DOE RU from early production to mid-1999, including ultimate use and disposition.
- Identify the characteristics and contaminants in the major U streams, specifically, the  $^{99}\text{Tc}$ , Np, Pu or other isotopic content of concern to worker or public health and safety.
- Conduct site mass balance activities sufficiently thorough to identify any significant implications for potential personnel exposure or environmental contamination.

PGDP processed RU from initial startup in fiscal year 1953 through fiscal year 1989 (excluding fiscal years 1965 through 1968, 1976A, and 1977 through 1985). Sources of the RU received were:

- Oxides from Hanford for conversion to  $\text{UF}_6$  at PGDP – 74,479 MTU (MTU – 1000 kilograms of U)
- Oxides,  $\text{UF}_6$  feed, and ash scrap from the Oak Ridge Gaseous Diffusion Plant (ORGDG) that originated from Hanford, Savannah River, or miscellaneous foreign and domestic sources - 11,684 MTU.
- Oxides from Savannah River for conversion to  $\text{UF}_6$  at PGDP - 9231 MTU
- Oxides,  $\text{UF}_6$  feed,  $\text{UF}_4$  and scrap ash from FMPC - 4055 MTU
- $\text{UF}_6$ ,  $\text{U}_3\text{O}_8$  and  $\text{UF}_4$  feed from miscellaneous foreign and domestic sources - 1842 MTU

These RU receipts total approximately 101,291 MTU. Approximately 89,064 MTU (from accountability reports, material codes 24 and 30) of this material was fed to the PGDP cascade from feed plant production and  $\text{UF}_6$  an additional 1,077 MTU (primarily UK Material) was also fed to the cascade under a different material code. This number is not reflected in total fed. Shipments of  $\text{UF}_6$ , oxides,  $\text{UF}_4$ , ash and miscellaneous material have totaled 3643 MTU. 1267

MTU remains on inventory. The MUF (material-unaccounted-for) of 6240 MTU may be due to loss of identity as RU material or disposal as waste.

PGDP also shipped U materials that were contaminated with TRU and fission product materials. PGDP processes removed TRU materials from the UF<sub>6</sub> process material and Np was also recovered for use in other DOE programs. The primary shipments of materials containing TRU or fission product materials consisted of the following:

- UF<sub>6</sub> product containing <sup>99</sup>Tc and non-detectable quantities of Pu and Np shipped to ORGDP- 85,230 MTU
- UF<sub>6</sub> product containing <sup>99</sup>Tc and non-detectable quantities of Pu and Np shipped to Portsmouth Gaseous Diffusion Plant (PORTS) – 119,167 MTU
- Enriched feed plant UF<sub>6</sub>, UF<sub>4</sub>, oxides, and ash scrap to FMPC- 9,070 -MTU

Receipts, shipments, plant processes, and available analytical data have been evaluated to determine the quantities of Pu, Np and <sup>99</sup>Tc related to plant activities. Based on these evaluations, the following balance estimates have been determined:

<b>Material</b>	<b>Received</b>	<b>Recovery Operations (To ORNL)</b>	<b>MUF*</b>	<b>Shipped or Returned to Others</b>	<b>Current Inventory</b>
<b>Pu</b>	300	17	210	70	3
<b>Np</b>	22,900	4,300	14,125	3,525	950
<b><sup>99</sup>Tc</b>	684,400	25000	422,048	237,352	280

\* MUF, Material Unaccounted For; Assumed lost to environment (releases, burial, and liquid discharges)

- All amounts listed above are in grams.
- Current inventory includes waste and amounts in unfed RU UF<sub>6</sub>.

Data estimates differ from those proposed by Smith chiefly because of examination by source and type rather than feed to the cascade as NMC&A code 524 and 624 material. The approach used in the estimates produced in this document resulted in a slight, but insignificant, difference in the total amount of impurities received.

The RU processed at PGDP presented the potential for worker exposure greater than that attributable to U alone. Feed plant processing of oxides from Savannah River and Hanford tended to concentrate both Pu and Np in the ash byproduct. Feed plant workers were susceptible to elevated exposures due to the work environment. Fluorinated Pu and Np from the feed plant would further concentrate in UF<sub>6</sub> cylinders and at the cascade feed points and lower stages. Worker exposures were potentially elevated during cascade upgrade projects when equipment was removed that had TRU deposits.

# **RECYCLED URANIUM MASS BALANCE PROJECT PADUCAH GASEOUS DIFFUSION PLANT SITE REPORT**

## **1.0 Paducah Gaseous Diffusion Plant Uranium Mass Balance Project**

### **1.1 Project Overview**

The overall objective of the mass balance project is to identify where RU could have created an exposure hazard to the workers and an estimation of the numbers of workers potentially exposed. The project also includes elements to identify significant contamination to the environment. In order to meet these objectives, the historical flow and processing of RU required evaluation to meet the DOE project plan objectives:

- Identify the mass flow of DOE RU from early production to mid-1999, including ultimate use and disposition.
- Identify the characteristics and contaminants in the major U streams, specifically, the  $^{99}\text{Tc}$ , Np, Pu or other isotopic content of concern to worker or public health and safety.
- Conduct site mass balance activities sufficiently thorough to identify any significant implications for potential personnel exposure or environmental contamination.

### **1.2 Purpose and Scope**

The purpose of the PGDP mass balance project is to identify the historical mass flows and characteristics of irradiated RU generated and processed by DOE over the last 50 years and subsequently shipped to and from PGDP. This information will enable the DOE to assess the potential for worker exposure and environmental contamination at PGDP attributable to the TRU isotopes of Pu, Np, and the fission product  $^{99}\text{Tc}$ . These contaminants are known to have been present in the U recycled from DOE reactor programs and other sources.

All U bearing materials processed by DOE reactors and foreign source material processed by reactors and subsequently shipped to PGDP are within the scope of this project. Materials of concern for PGDP originated at the DOE Hanford and Savannah River sites and foreign source reactors. Some of the material originating at Hanford and Savannah River may have been shipped to PGDP through the FMPC Feed Materials Production Center (FMPC) and ORGDP. The U received at PGDP was either enriched or depleted and consisted of the following chemical forms: Uranium fluorides ( $\text{UF}_6$ ,  $\text{UF}_4$ ), uranium oxides ( $\text{UO}_2$ ,  $\text{UO}_3$ ,  $\text{U}_3\text{O}_8$ ), and scrap compounds resulting from feed plant production. These same material types, when shipped from the PGDP site are also within the scope of the mass balance project. These types of materials were shipped to the following major sites: Oak Ridge Gaseous Diffusion Plant (ORGDP), Portsmouth Gaseous Diffusion Plant (PORTS), and FMPC.

### **1.3 Project Implementation Strategy**

In order to facilitate the implementation of the project, PGDP assembled a team to identify and evaluate historical documentation related to the shipments and receipts of RU. RU shipments, receipts, cascade feed and inventory were identified through review of Nuclear Material Control and Accountability (NMC&A) Monthly Balance Reports (MBRs), historical NMC&A data reports recorded on microfiche, Nuclear Materials Management and Safeguards System (NMMSS) reports, and NMC&A organization inventory database reports. Data from these sources for the period July 1952 through 1999 were reviewed and summaries can be found in the appendices to this report.

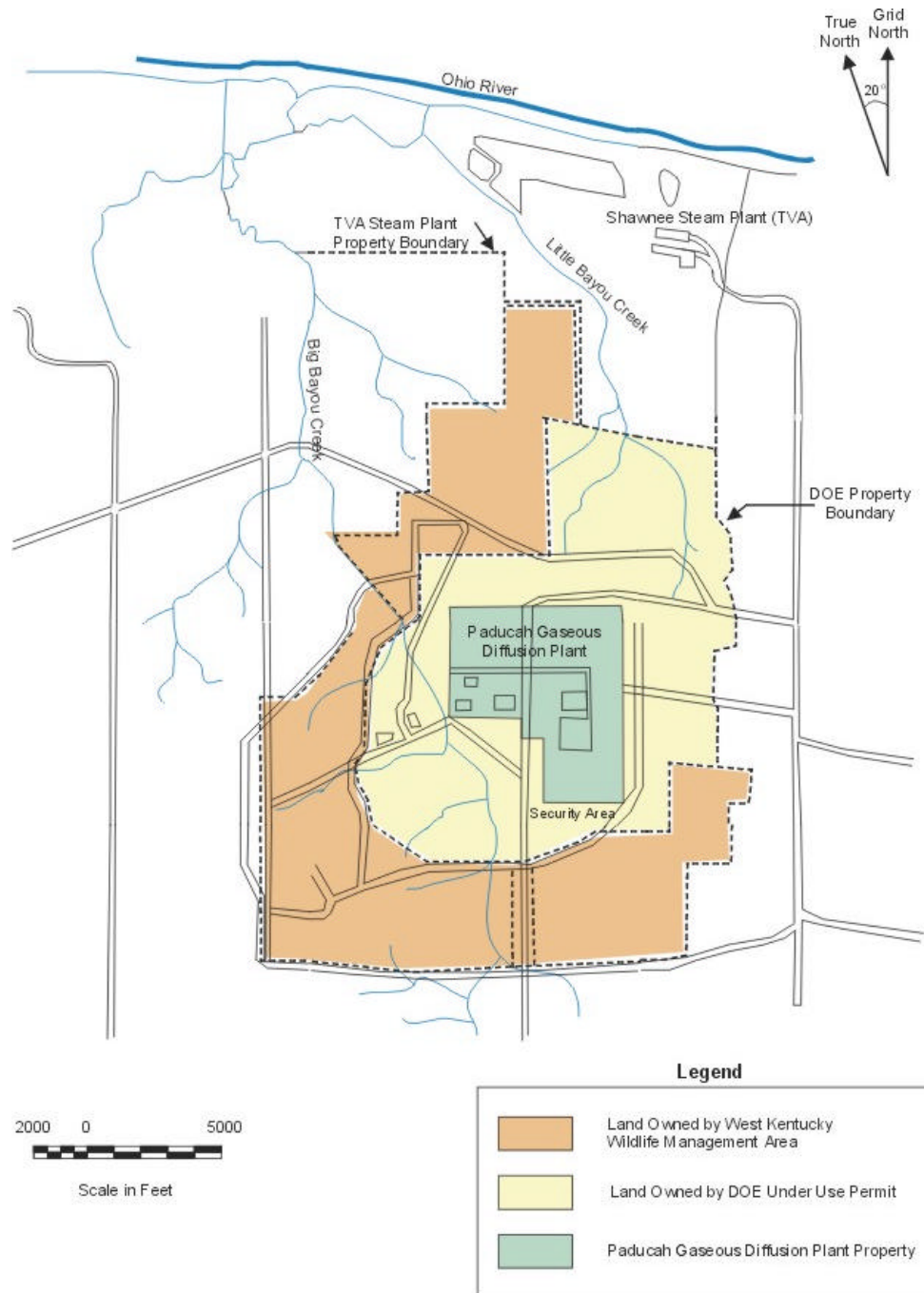
Limited analytical data documenting TRU and fission product contamination were identified for the RU shipments, receipts, feed plant processing and cascade feeds. This data is derived from of a mix of laboratory reports, historical documents on RU (e.g., the Smith Report), and environmental reports. The application of this data to determine the quantities of TRU and FP involved is discussed in chapter 4 and 5 of this report.

## **2.0 Site Historical Overview**

### **2.1 Site Description**

The PGDP is located in McCracken County, Kentucky, approximately 10 miles west of the City of Paducah and 3 miles south of the Ohio River. The site occupies 3,425 acres, 748 of which are within a security fence, and contains U enrichment process equipment and support facilities. The mission of the plant is to "enrich" U for use in domestic and foreign commercial power reactors. Enrichment involves increasing the percentage of  $^{235}\text{U}$  in the material ( $\text{UF}_6$ ) used for creating reactor fuel.  $^{235}\text{U}$  is highly fissionable, unlike the more common isotope uranium-238. The PGDP enriches the  $\text{UF}_6$  from roughly 0.7 percent  $^{235}\text{U}$  (natural uranium) to about 2.0 percent  $^{235}\text{U}$ . Over its operating lifetime, PGDP has processed more than one million tons of U. The process of enriching U at PGDP involves heating  $\text{UF}_6$  into a gas, which is in turn fed through a series of diffusion stages; PGDP has more than 1,800 diffusion stages. The diffusion process generates enriched U product and depleted by product referred to as tails. The product (or slightly enriched material) is shipped to the Portsmouth Gaseous Diffusion Plant in Ohio, where it is normally enriched to 3 to 5 percent  $^{235}\text{U}$ . The tails (depleted material), which typically containing less than 0.4 percent  $^{235}\text{U}$ , remain on site in cylinders. DOE is the site "landlord," owning the land and physical plant. Since July 1993, all of the enrichment process buildings and various enrichment support facilities have been leased to the United States Enrichment Corporation (USEC) for the production of low enriched uranium. Because of past plant operations, hazardous substances, wastes, or constituents were released into the environment and have required the DOE to thoroughly perform various investigation and remediation activities. In order to adequately address the environmental contamination at the PGDP, DOE established the Environmental Restoration (ER) Program. The on-going DOE ER program activities for the PGDP are: operation of the Northwest plume and the Northeast plume groundwater treatment systems; operation of a permitted solid waste contained landfill; operation of waste treatment and storage facilities; waste characterization and disposal; maintenance of non-leased roads, grounds, and facilities; surveillance and maintenance of depleted  $\text{UF}_6$  cylinders; construction of new cylinder yards; maintenance of closed landfills and burial sites; environmental monitoring; and environmental restoration. Bechtel Jacobs Company has been the management and integrating contractor for DOE since April 1998. Bechtel Jacobs Company uses subcontractors to accomplish much of the scope of its contract with DOE. Figure 2.1-1 shows major boundaries and features of the site.



**Figure 2.1-1, Map of Paducah Gaseous Diffusion Plant, Major Boundaries and Features**

The Energy Policy Act of 1992 mandated that a new government corporation (The United States Enrichment Corporation or USEC) would take over U enrichment production at the Paducah and Portsmouth Gaseous Diffusion Plants. DOE retained ownership of the plants, but leased the essential facilities to USEC. USEC leased the U enrichment production facilities from DOE on July 1, 1993, and contracted with Lockheed Martin Utility Services, Inc. as its operating and maintenance contractor. USEC became a private corporation in July 1995 and continued to use LMUS as its contractor. USEC assumed direct operation of the enrichment activities in May

1999. The NRC provides regulatory oversight of USEC activities. The Occupational Safety and Health Administration (OSHA) regulates USEC and DOE occupational worker safety and health, and the Commonwealth of Kentucky and the U.S. Environmental Protection Agency (EPA) regulate USEC and DOE environmental activities. Facilities leased by USEC consist of process buildings, electrical switchyards, a steam plant, a water treatment facility, a chemical cleaning and decontamination facility, and maintenance and laboratory facilities.

### **Site History**

In August 1950, the United States government determined that it would need to double the capacity of domestic fissionable materials production at the Oak Ridge, Tennessee ORGDP Gaseous Diffusion Plant. The Atomic Energy Commission (AEC) selected a plant option consisting of 400 stages modeled after the K-31 facility (which would become C-331 at the Paducah Plant) and one with 480 stages, twice the size of the Oak Ridge K-31 stages (which would become C-333 at the Paducah Plant). Based on a decision to disperse the major portions of the new production capacity, eight areas were identified as candidate locations for the plant, all in the southeastern United States. From the application of additional criteria, three sites were identified: the Kentucky Ordnance Works (KOW) at Paducah, the Louisiana Ordnance Plant at Shreveport, and the Longhorn Ordnance Works at Marshall, Texas. From these, the AEC approved, on October 18, 1950, the KOW site as the location for the new gaseous diffusion plant.

PGDP construction spanned 1951 through 1956 and was conducted in two phases. Construction of the first phase began January 2, 1951, and included the following process and production facilities: C-331 and C-333, the gaseous diffusion process buildings; C-410/420, UF<sub>6</sub> Feed Plant; C-310, Purge and Product Withdrawal Building; C-315, Surge and Tails Withdrawal Building; and C-300, Central Control Building. On January 6, 1951, the Tennessee Valley Authority (TVA) began construction of the four-unit Shawnee Steam Plant near the PGDP on the Ohio River to provide a portion of the electricity required to operate the new gaseous diffusion plant. On February 15, 1951, Electric Energy, Inc., began construction of the Joppa Steam Plant, in Joppa, Illinois, also to provide electricity to PGDP. Authorization to proceed with the second phase of PGDP construction was received on July 15, 1952. Two additional enrichment facilities, C-335 and C-337, were added, and construction was completed in 1956. Carbide and Chemicals Company (which became Union Carbide Corporation Nuclear Division) was named as the original site contractor based on the company's experience with gaseous diffusion operations at Oak Ridge, Tennessee. Carbide operated PGDP for the AEC, and its successor agencies, the Energy Research and Development Administration (ERDA) and DOE, until 1984, when Martin Marietta Energy Systems, Inc., won the contract through a competitive procurement.

Major milestones in PGDP's operations are summarized in Table 2.1-1, Plant Operations Affecting Np, <sup>99</sup>Tc, and Pu.

**Table 2.1-1, Plant Operations Affecting Neptunium, Technetium, and Plutonium Flow Streams**

October 1950	Paducah selected as site for new gaseous diffusion plant
January 1951	Construction begins
July 1952	First U received at Paducah
September 1952	Cascade buildings C-331 and C-333 begin operation
November 1952	First product withdrawn
April 1953	C-400 cleaning building activated
July 1953	First reactor tails (RT) UF <sub>6</sub> fed to the enrichment cascade, after conversion in Oak Ridge
July 1953	Feed plant started – converted RT uranium trioxide (UO <sub>3</sub> ) to UF <sub>6</sub>
1953-1954	Filter press sludge containing U decay products and Np, Pu, and some fission products were buried with fluoride wastes
April/July 1954	C-335 and C-337 begin operation
1955	Process started to dissolve feed plant ash, recover U and discard impurities, including <sup>99</sup> Tc, Np, and Pu
August 1956	C-420 expansion to feed plant completed
December 1956	C-340 UF <sub>6</sub> to uranium tetrafluoride (UF <sub>4</sub> ) conversion process on stream
January 1957	C-340 U derby production started
1957	Presence of Np identified in reactor tails feed to cascade. Raffinate from selected ash processing sampled and analyzed.
1957	C-404 Holding Pond converted to solid radioactive waste burial area. Liquid wastes channeled to Little Bayou Creek; land along creek bought to provide DOE ownership from the plant to the Ohio River
November 1958	Production begun to recover and purify <sup>237</sup> Np for other AEC programs
April 1960	Production begun for the recovery of <sup>99</sup> Tc for other AEC programs
September 1961	Magnesium fluoride traps for <sup>237</sup> Np and <sup>99</sup> Tc installed in the Feed Plant UF <sub>6</sub> system
March 1962	Explosion and fire in C-340 Metals Plant; one fatality
March 1962	Np recovery stopped; the 4,289 grams <sup>237</sup> Np recovered in this process and about 1000 grams recovered by Oak Ridge National Laboratory (ORNL) satisfied the needs
December 1962	Explosion and fire in C-337

**Table 2.1-1, Plant Operations Affecting Neptunium, Technetium, and Plutonium Flow Streams**

January 1963	Magnesium fluoride ( $\text{MgF}_2$ ) traps installed in the purge cascade near the product withdrawal to reduce $^{99}\text{Tc}$ and Np in product and to reduce $^{99}\text{Tc}$ in vent gases; traps not serviced regularly from the shutdown of the Feed Plant in 1964 until 1975; after this the Np can go down stream
June 1964	Feed Plant shut down for more than 4 years. RT feed to cascade ceased
April 1968	Radiation overexposure to 2 maintenance workers
July 1968	Feed Plant prepared for renewed operation
March 1970	Wet recovery of ash and other materials with concentrated Np, $^{99}\text{Tc}$ , and Pu ceased; materials stored for later recovery or shipment
March 1973	Second CIP/CUP started
FY 1973	About 65% of all cascade feed was from recycled RT uranium
October 1973	C-340 U derby production discontinued
January 1975	NRC and ERDA assume regulatory responsibilities for AEC activities
September 1975	Last RT feed of this era vaporized to cascade
May 1977	Feed Plant shut down
July 1977	Waste water treatment plant began operation; major waste water stream changed from plant ditch to Little Bayou Creek to waste water treatment plant lagoon that overflows to Big Bayou Creek
October 1977	DOE assumes regulatory responsibilities from ERDA
January 1978	Explosion and fire in C-315, Tails Withdrawal Facility
September 1981	Second CIP/CUP completed
April 1984	Martin Marietta replaces Union Carbide as site operating contractor.
June 1986	Discovery of major TCE leak from C-400 cleaning facility
August 1988	Discovery TCE and Tc in offsite private residential wells
1992	USEC established by Energy Policy Act of 1992
July 1993	USEC leases enrichment production facilities from DOE; Lockheed Martin Utility Services becomes USEC operations and maintenance contractor; Lockheed Martin Energy Systems continues as management and operating contractor for DOE
November 1996	NRC grants certificate of compliance for enrichment operations
March 1997	Regulatory oversight of enrichment transferred from DOE to NRC

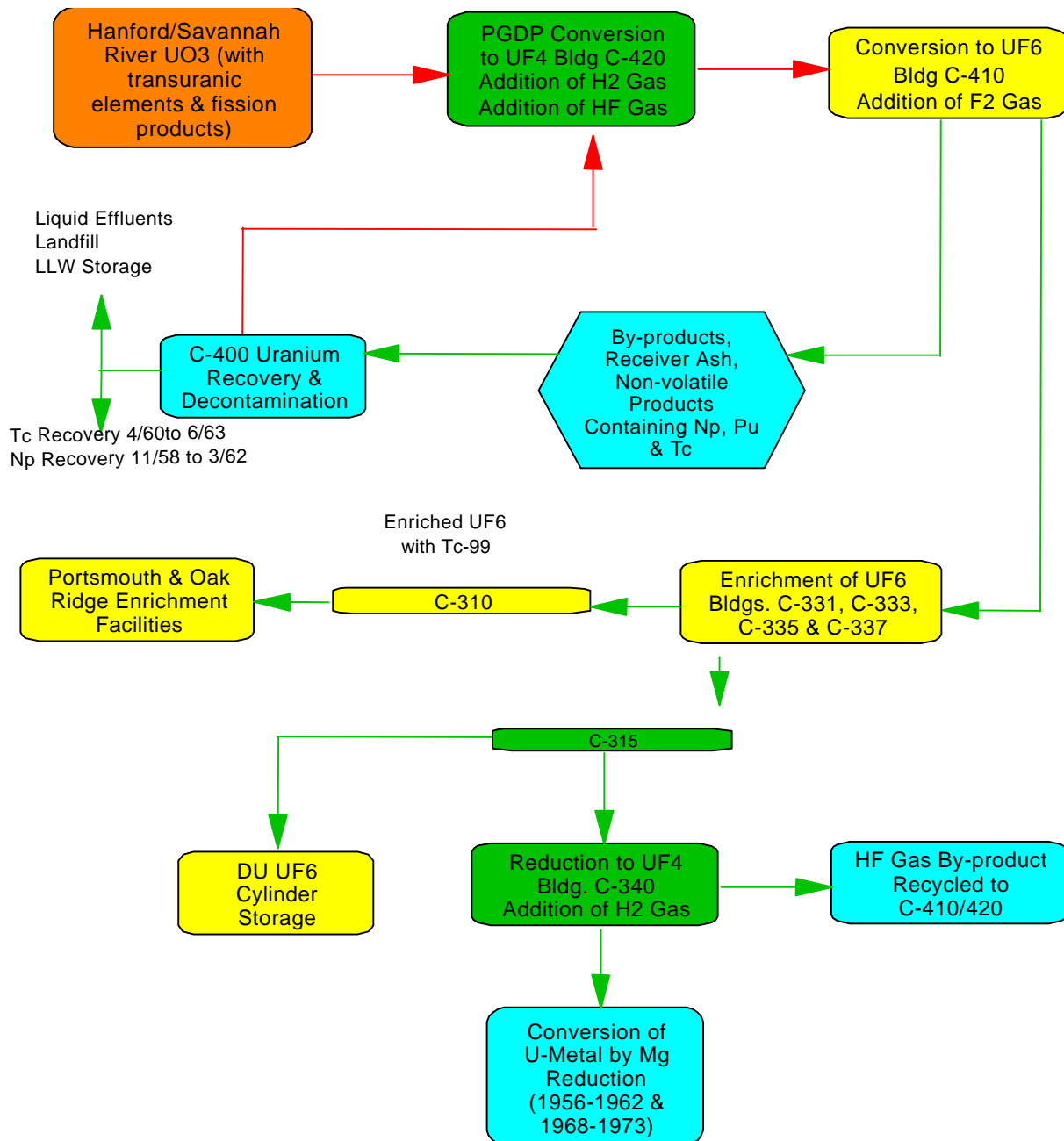
**Table 2.1-1, Plant Operations Affecting Neptunium, Technetium, and Plutonium Flow Streams**

April 1998	Bechtel Jacobs Company awarded DOE management and integration contract
July 1998	USEC becomes private corporation
May 1999	USEC takes over direct operation of enrichment activities

The purpose of the gaseous diffusion plant has been and continues to be the enrichment of U, initially for military applications and subsequently for commercial nuclear reactor fuel. PGDP enriches feed material in the form of UF<sub>6</sub> gas from approximately 0.711 percent <sup>235</sup>U to UF<sub>6</sub> with one to about two percent <sup>235</sup>U. The enriched product from PGDP was sent to other DOE gaseous diffusion plants at Portsmouth, Ohio, or Oak Ridge, Tennessee, for further enrichment. Originally most UF<sub>6</sub> feed material came from the depleted tails produced during normal diffusion operations at PGDP and from Oak Ridge and Portsmouth. From 1952 through 1977, UF<sub>6</sub> feed material was also produced from uranium trioxide or UO<sub>3</sub> (called "yellowcake") at PGDP in Buildings C-410 and C-420; this feed material was supplied by sources such as El Dorado Mining and Refining, Mallinckrodt Chemical Works, and General Chemicals (now Allied Chemical) and comprised less than 10 percent of the UF<sub>6</sub> fed to the cascade. From 1953 through 1964 and intermittently from 1968 through 1977, the Feed Plant also produced UF<sub>6</sub> from UO<sub>3</sub> from spent reactor fuel processed at the Hanford and Savannah River sites. After 1977, all feed came in the form of UF<sub>6</sub> from outside sources such as Oak Ridge, Portsmouth, and Allied Chemical. Detailed information showing U receipts and shipments are provided in Appendix A.

Although natural U is not a highly radioactive material, it is toxic, both chemically and radiologically, when inside the body. The U exposure pathway of greatest hazard at PGDP was inhalation of U dust. Feed material was made from production reactor tails from 1953 until 1964, and intermittently from 1968 to 1977. The percentage of PGDP cascade feed material from reactor tails averaged 17 percent during the periods this material was used, ranging from 3 percent in 1975 to 65 percent in 1973. Detailed information showing the use of reactor tails in the cascade is provided in Appendix B. Processing of UO<sub>3</sub> into UF<sub>6</sub> was accomplished in three steps: reduction, hydro-fluorination, and fluorination (see Figure 2.1-2).

Figure 2.1-2, Historical Uranium Enrichment Process



Reduction involved transforming  $\text{UO}_3$  into  $\text{UO}_2$  using hydrogen gas. Hydro-fluorination of  $\text{UO}_2$  into  $\text{UF}_4$  (commonly referred to as "green salt") was accomplished by adding anhydrous hydrofluoric acid (HF). Fluorination was conducted in C-410 using heated elemental fluorine gas in tower reactors. The first two steps were performed in C-410 on vibration tray reactors (shaker trays) from 1953 to 1956. In 1956, due to frequent equipment failures, spills, leaks, and the increased demand for feed, Building C-420 (commonly called the "green salt" plant) was completed and "green salt" production at C-410 was phased out. In C-420, the reduction was performed in two-stage fluidized bed reductors; the hydro-fluorination was performed in three sets of horizontal screw reactors or in a two-stage fluidized bed hydro-fluorinator. High radiation areas existed near the fluorination towers and ash receivers. Respirators were specified for most work activities. The Feed Plant was shut down in 1977.

## 2.2 Key Uranium Processing Facilities

The major facilities at PGDP are:

- C-331, C-333, C-335, and C-337 – Gaseous Diffusion Process Buildings
- C-410/420 –  $\text{UF}_6$  Feed Plant (inactive)
- C-300 – Central Control Building
- C-310 – Purge and Product Withdrawal Building
- C-315 – Surge and Tails Withdrawal Building
- C-340 – Metals Plant (inactive)
- C-400 – Decontamination and Cleaning Building

The main process buildings at PGDP (C-331, C-333, C-335, and C-337) contain the "cascades," which are a series of compressor and converter stages and supporting equipment arranged in units and cells that progressively enrich the  $\text{UF}_6$  in its gaseous form. Enrichment occurs as the  $\text{UF}_6$  passes through barriers in the converters allowing isotopes of lower molecular weight to pass through. The series of converters results in two streams of  $\text{UF}_6$ : one of progressively higher-percentage  $^{235}\text{U}$  that moves to the product withdrawal station in C-310, and one of progressively higher-percentage  $^{238}\text{U}$  that moves toward the tails withdrawal station in C-315. Both the enriched product and the depleted tails are fed as liquid into cylinders and allowed to cool until solid. The product is shipped to Portsmouth for further enrichment. The depleted material is either re-fed to the cascade or stored on site.

In 1957, the presence of Np and  $^{99}\text{Tc}$  was documented at PGDP, and between 1959 and 1966 numerous studies related to the behavior, health effects, and controls for these elements were conducted by PGDP and the AEC (Phase II Investigation). The concentration of TRUs, such as Np and Pu, and FP, such as  $^{99}\text{Tc}$ , in the reactor tails material was very small, estimated at approximately 0.2 parts per million (ppm) Np, 4 parts per billion (ppb) Pu, and 7ppm  $^{99}\text{Tc}$ . However, these elements are much more hazardous than natural U and were concentrated by during processing at certain specific locations, presenting increased hazards to certain workers. Np has a specific activity up to 2,000 times greater than an equivalent amount of U, depending on the level of enrichment. Pu is significantly more radioactive than Np, but constituted a lesser hazard because it was present in much lower concentrations. Both Pu and Np are significant radiation hazards if inhaled or ingested.  $^{99}\text{Tc}$  is primarily a beta emitter with a higher specific activity than U, and is highly mobile in groundwater.

Approximately 25 percent of the Np in the feed material remained in the Feed Plant as dust or ash. Approximately 50 percent remained in cylinder heels after feeding, and approximately 25 percent was vaporized in the cascade.  $^{99}\text{Tc}$  tended to migrate to the top of the cascade, and much was drained off into the product or vented to the atmosphere. In 1958, a Np recovery process was initiated in C-400 to recover Np from the fluorination ash and cylinder heels. A program to recover  $^{99}\text{Tc}$  from the cylinder wash water and raffinate (e.g., solvents) from Np recovery operations began in April 1960. In September 1961, magnesium fluoride pellet traps were installed in the Feed Plant to capture Np and  $^{99}\text{Tc}$ ; in January 1963, similar traps were installed at the C-310 product withdrawal stations. By March 1962, Np recovery operations had ended, and in June 1963,  $^{99}\text{Tc}$  recovery operations also ceased. A different  $^{99}\text{Tc}$  recovery process was initiated in the mid-1970s to remove  $^{99}\text{Tc}$  from aqueous waste streams for the purpose of environmental protection. (KY-L-936  $^{99}\text{Tc}$  and TRU Summary)

Before the mid-1970s, a complex U recovery process was operated in C-400 for separating U from waste and scrap materials, concentrating and converting it to an oxide. The U recovery system was not leak-tight, and leaks were common. However, steps were taken to control operators' exposure to process materials. Routine surveys were conducted to monitor the concentration of radioactivity on surfaces and in the air in C-400, and the health physics staff recommended changes in work practices based on the results of these surveys. In the mid-1970s, the solvent extraction process for U recovery was replaced with a simpler precipitation and filtration process. The filtrate, containing low concentrations of radionuclides, was discharged to the environment via the C-400 drains. Sludges and filter cake were processed at PGDP for U recovery or sent to FMPC for recovery.

From December 1956 through December 1962 and from January 1968 through October 1973, PGDP produced  $\text{UF}_4$  and U metal in C-340 for weapons uses.  $\text{UF}_4$  Production continued until 1977. The U metal production process involved reducing  $\text{UF}_6$  (normally from the tails cylinder) to  $\text{UF}_4$  by combining it with hydrogen in a heated tower. The  $\text{UF}_4$  was mixed with magnesium and fed into lined firing reduction vessels (commonly referred to as "bombs"), placed in furnaces, and heated until it fired into a metal ingot, called a "derby." The derbies were removed from the bomb, cleaned, cut, and shipped to Oak Ridge.

### 2.2.1 Feed Plant Operations

In order to enrich the U in the cascades, the feed product has to be in the form of  $\text{UF}_6$ . PGDP currently receives  $\text{UF}_6$  directly from various customers. Before 1976, however, much of the U was received from the various ore processing refineries and reactor U recovery facilities (Savannah River and Hanford) in the form of  $\text{UO}_3$ , also commonly known as "yellow powder" or "yellowcake." This material was then converted to  $\text{UF}_6$  by a three-step reaction process in the C-410/-420 feed plant, which operated from July 1953 through June 1964 and from July 1968 through June 1977. In the first step, the  $\text{UO}_3$  was reduced to uranium dioxide ( $\text{UO}_2$ ) by reacting with hydrogen ( $\text{H}_2$ ). The  $\text{UO}_2$  was then reacted with HF to produce  $\text{UF}_4$ , also commonly known as "green salt." The  $\text{UF}_4$  was finally converted to  $\text{UF}_6$  with fluorine ( $\text{F}_2$ ).

Operating and maintenance practices in the feed plant were generally consistent with accepted industrial practices at the time, although the work environment was harsh. From the feed plant startup in 1953 until 1956, there were three lines for processing  $\text{UO}_3$  to  $\text{UF}_6$  located in C-410. In each line, the first two steps of feed production ("green salt" production) were conducted on vibrating tray reactors (shaker trays): a 15-foot-long tray for  $\text{UO}_2$  production and two 40-foot long trays for  $\text{UF}_4$  production. Each line contained a fluorination tower for converting "green salt" to  $\text{UF}_6$  gas. Unexpected harmonic stresses on the trays resulted in frequent failures of the



trays and bellows, with subsequent spills and leaks of U powders and gases, thereby contributing to the harsh working environment. These failures, combined with increased demand for feed, resulted in the addition of five more fluorination towers and the C-420 "green salt" feed plant, which replaced the shaker trays with screw reactor and fluid bed technologies. These technologies also had their share of problems. Room temperatures in the feed plant were usually in excess of 100 degrees Fahrenheit, noise levels were high, and leaks in all systems were common throughout the life of the plant.

Exposure to U powder dusts was prevalent in both operations and maintenance activities (Phase II Investigation). For example, plugging of conveyers, hoppers, and screws with  $\text{UO}_3$  or  $\text{UF}_4$  routinely required physical agitation with sledgehammers or metal rods. In many cases, shear pins or chains on the associated drive mechanisms broke, requiring operations personnel to clean the product out of the jammed equipment and maintenance personnel to disassemble and repair the equipment.

In certain areas of the feed plant, U daughter products, TRUs, or FP tended to concentrate. These areas included the plant dust collection systems, the fluorination towers, and the ash receivers downstream of the fluorination towers. Vacuum and ventilation system bag rooms exposed workers to fine particle dust containing appreciable concentrations of the impurities. The impurities deposited out on the inside of the fluorination towers, making them radiation areas. The ash resulting from the fluorination of the  $\text{UF}_4$  contained the most radioactive impurities and was sometimes in the form of small particulates. As a result, the ash receivers provided one of the highest potentials for exposures to workers. Ash receivers were hot and fuming, and at least one full ash receiver usually needed changing out each shift. In addition, plugging of towers with ash frequently required physically challenging manual cleanout, putting workers in close proximity to the towers and the ash plugs for long periods of time. Respirators were typically required for most of this work.

### **2.2.2 Cascade Operations, Feed, and Withdrawal**

The cascades generally operated below atmospheric pressure, and therefore, any leakage results in air flowing into the process. The cylinder feed system and the product withdrawal system operated above atmospheric pressure. Any leakage in these areas resulted in process gas venting into and contaminating the surrounding atmosphere. In addition, the "heels" in empty cylinders brought to the withdrawal areas or removed from the feed areas were a source of penetrating radiation for the workers. Cylinder heels are composed of non-volatile corrosion products, U salts and oxides, and residual TRU and U daughter product compounds remaining in the cylinders when  $\text{UF}_6$  is fed to the cascade. Without the self-shielding effects of the U in a full cylinder, the empty cylinders produced appreciable gamma fields. Since cylinders were re-used for five-year periods between cleaning and testing, heels in some cylinders accumulated significant radiation sources.

During the 1950s,  $\text{UF}_6$  gas was pressurized for feeding to the cascade by heating the cylinders in warm water baths; the water baths had minimal engineered safety features. In November 1960, a cylinder was valved into the cascade before the water bath was fully heated, resulting in backflow into the cylinder from the cascade and an overfill condition. When the inappropriate valving was discovered, the cylinder isolation valve was closed. As the water bath continued to heat the cylinder, the cylinder over pressurized, rupturing the cylinder and releasing approximately 6,800 pounds of U (Phase II Investigation).

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In the early 1960s, the water baths were replaced with autoclaves, located in Buildings C-333-A and C-337-A, with each building containing several autoclave feed stations. Prior to connection to the cascade, each UF<sub>6</sub> cylinder was inspected for damage and confirmed to be safe for use. If a cylinder was found to be defective, it was tagged and moved aside for special handling. Following inspection, a heat traced copper pipe (pigtail) was attached to the cylinder valve and to a corresponding connection at the autoclave station, the cylinder valve opened, the autoclave closed, and the various alarms tested. Once the connection integrity and feed path clearance were confirmed, steam heat was initiated to vaporize the UF<sub>6</sub> and began feeding it to its corresponding assay point in the cascade. A UF<sub>6</sub> release within an autoclave would actuate an automatic emergency shutdown and autoclave isolation to protect workers and the environment.

Enriched and depleted UF<sub>6</sub> gas was withdrawn in Buildings C-310 and 315, respectively. Product (enriched UF<sub>6</sub>) and tails (depleted UF<sub>6</sub>) were withdrawn from the cascade by pumps that discharged through a condenser, piping, and cylinder pigtails to the intended receiving UF<sub>6</sub> cylinder. Product cylinders were to be filled to no more than 95 percent (liquid) of capacity. Those that were overfilled were tagged and subject to special handling to resolve the overfilled condition. UF<sub>6</sub> cylinders still containing liquid could not be transported around the site without special consideration. Before solid UF<sub>6</sub> cylinders were moved to storage, they were "burped" of light gases through sodium fluoride (NaF) traps.

Portions of the product withdrawal system operated at approximately 30 psig. As a result, small leaks in this area released enriched process gas into the room atmosphere and provided a higher potential for an intake. Air monitor sampling indicated moderately high activity readings for the withdrawal room from initial operations up through the early 1960s (Phase II Investigation). Subsequent increased attention to repairing leaks and improving the ventilation systems led to low activity readings in the room by 1964. Other than a few specific high readings due to leaks, general area air monitoring samples remained low.

Accidental UF<sub>6</sub> releases during the connection and disconnection of cylinders was one of the leading causes of individuals reporting to the dispensary for medical attention in 1953, according to a PGDP quarterly report (Phase II Investigation). UF<sub>6</sub> releases often occurred when burping recently filled UF<sub>6</sub> cylinders. Workers generally wore full-face respirators during this activity and received monthly bioassays. In one instance, a worker attempted to move a product cylinder that was still connected to its pigtail, resulting in a major UF<sub>6</sub> release (Phase II Investigation). Workers reportedly received skin burns while attempting to isolate the release. Interlocks were subsequently added to prevent a recurrence.

### **Puffs**

Puffs are minor releases of UF<sub>6</sub> from process gas equipment and were a common occurrence, despite efforts to minimize the amount of material available for release. Puffs could occur during routine maintenance activity or pigtail disconnection. Workers often determined when a job might produce a puff and, therefore, whether a respirator should be worn based on prior experience (Phase II Investigation).

### **Jetting**

Jetting is a term used to describe the purging of isolated process gas system equipment of UF<sub>6</sub> and HF by introducing dry air or nitrogen and removing the resulting gaseous mixture with the process building purge jets. Each jet drew suction on its process building evacuation header, which consisted of a two-stage Venturi supplied with 100-pound air, and discharged the resulting

gaseous mixture to the environment from an unmonitored open pipe on the process building roof (Phase II Investigation). The jets were intended to evacuate atmospheric air from isolated process gas system equipment in preparation for startup and the introduction of  $\text{UF}_6$  and for performing HF sweeps of isolated process gas system equipment. These HF sweeps were performed once the  $\text{UF}_6$  concentration had been reduced below 10ppm ( $\text{UF}_6$  negative) in preparation for opening the process gas system for maintenance, inspection, or parts retrieval. Assuming that the jets were only used as prescribed after a satisfactory  $\text{UF}_6$  negative was achieved, less than one-fifth of a pound of  $\text{UF}_6$  was available for release to the environment from a single cascade cell each time. The number and frequency of these authorized releases is not known.

Other jetting activities included the use of the jets at night to accelerate the attainment of an adequate  $\text{UF}_6$  negative to support a planned opening of isolated process gas equipment. Depending on the pressure, temperature, and concentration of  $\text{UF}_6$  in a cascade cell when jetting was initiated, and assuming that the concentration had been reduced by at least one-tenth through purging and evacuation pumps, up to several thousand pounds of  $\text{UF}_6$  could still have been available for release to the environment from a single cascade cell (Phase II Investigation). As with normal jetting, the  $\text{UF}_6$  gas would hydrolyze with moist air to form  $\text{UO}_2\text{F}_2$  powder and HF gas. The number and frequency of these inappropriate releases were not determined during this investigation. Plant management did not authorize this process. Where discussed in the procedures, the use of jetting was limited to static or sweep purging of isolated process gas equipment after a satisfactory  $\text{UF}_6$  negative had been achieved and confirmed by sampling. Procedures from the late 1980s and 1990s do not address jetting at all, relying instead on evacuated surge drums and wet air pumps to perform HF static and sweeping purges with essentially no release of  $\text{UF}_6$  to the environment. Additionally, the jets were physically blanked off during this period.

### **2.2.3 $\text{UF}_4$ and Metal Production (C-340)**

Along with the enriched U produced at Paducah, the PGDP also produced U metal. These operations were conducted, following completion of construction in 1957, in a small complex of buildings on the eastern side of the PGDP known as C-340. In June 1962, operations were significantly scaled back. A second campaign began in 1967 and continued until 1977. From 1978 to 1982, the C-340 building served as a shipping point for  $\text{UF}_4$  “green salt”. After 1982, the C-340 building was used for utilities maintenance, training classes, security exercises, and prototype valve tests. In 1994, the C-340 building was fenced and locked, and it is currently in the decontamination and decommissioning program where it receives routine surveillance and maintenance. Decontamination and decommissioning activities are not anticipated to take place until after shutdown of the gaseous diffusion plant.

Metals production involved several steps, each with its own unique hazards. The first step in the process was powder production.  $\text{UF}_6$  process gas reacted with hydrogen in a heated tower to produce  $\text{UF}_4$  powder, (“green salt”, depleted U) and HF. The HF was vented from the tower through a collection system that condensed the HF to a liquid, which was stored in a tank. Periodically, the tank would be pressurized with nitrogen and the HF transferred to Building C-410 for use in feed production.

Army assault masks and respirators were normally available to operators and were required for many of the operations. Entries in operating instructions and reports from workers indicated that these requirements were not always followed or adequately stressed by foremen (Phase II Investigation). Consequently, operators in C-340 were frequently placed on restriction due to the

intake of U compounds, especially in the powder areas on the fifth and sixth floors of the tower building.

The reaction towers were a primary source of airborne U. Operating at pressures above atmospheric, any leak in the system could release fine dust and HF. The building had two vacuum systems (dust collectors), one for general cleaning and one for U, with hose ports that could be connected in many locations. These hoses were frequently placed near leak sources to minimize releases, but they were not always effective. Very early on, the general cleaning system became contaminated when it was used while the U system was shut down for maintenance (Phase II Investigation).

The UF<sub>4</sub> "green salt" fell out of the bottom of the tower into a series of hoppers and screws used for powder transfer (Phase II Investigation). It could then be placed into drums for sale or storage or sent to the next step. UF<sub>4</sub> was removed from the hoppers at the bottom of the reaction towers. Large amounts of airborne U dust were created by this operation. Within four months after startup, respirators were identified as being required for drumming operations (Phase II Investigation).

Metal was produced by reduction of the UF<sub>4</sub> to U metal with magnesium. The first step in the process was preparation of a reactor liner. Magnesium fluoride (MgF<sub>2</sub>) was placed in a steel shell and "jolted" (mechanically agitated) to pack the refractory and remove any voids. The next phase of the operation involved blending measured quantities of UF<sub>4</sub> (depleted U) with measured quantities of magnesium metal, and then pouring this mixture into the reactor liner. A refractory cap was then poured, and a lid was bolted to the top of the charged reactor. The charged reactor was then transferred to an induction furnace where it was heated to the point where the U reduction started.

The primary hazard associated with this part of the process was exposure to the airborne U dust during weighing, blending, and pouring. Respirators were required very early during the initial production operations. The reactors also presented a significant hazard from burning magnesium and molten U metal. A phenomenon described as "burnout" and "lid fires" occurred infrequently when the refractory liner was not correctly prepared. For example, burnouts occurred when the burning magnesium came in contact with the steel shell, melting through the shell and releasing the reactor contents into the furnace. Lid fires were similar, but occurred at the lid rather than the side of the shell. Such an occurrence led to a fatality in March 1962. Burnouts resulted in significant contamination of the furnace refractory and would normally require the entire furnace to be relined.

After the reactor was cooled, it was sent to the breakout area where the lid was removed, the shell was inverted, and the contents were dumped onto a grating, referred to as a "grizzly." The slag material, at this point a hard ceramic material, was broken into smaller pieces by beating it with a hammer. The pieces were dropped through a grating into a jaw crusher and sent to the slag plant. This operation was among the dirtiest jobs in C-340. Operators could become completely covered with black dust. Respirators were required and generally worn, although the extent of dust and contamination probably exceeded the protection they provided. The metal ingot, referred to as a derby, was freed from the slag and "roasted" to oxidize the surface and loosen any remaining slag. Loose oxides that fell from the derbies during roasting were collected, put in drums, and sent to a burial yard. After roasting, the derbies were cleaned by hand in a cleaning booth using power brushes and grinders to remove any remaining slag. The potential hazards for airborne contamination for this operation were similar to the breakout operation.

After cleaning, the derbies could be shipped directly or sawed into smaller shapes, depending on customer requirements. Derby sawing generated large amounts of U metal "saw dust," which burns readily in air. Consequently, saw dust was collected in drums of oil and kept covered. Despite these measures, U metal fires were common (daily or weekly) (Phase II Investigation), resulting in high levels of airborne uranium oxides.

The  $\text{MgF}_2$  reaction product remaining in the reactor was captured, crushed, ball milled, and then sized to be recycled as refractory. Although primarily a hands-off operation, it generated significant quantities of dust. Over time, the slag became contaminated with significant quantities of uranium oxides (several percent) that could have contributed to worker intakes. Reject slag (too small or too large) was collected in a hopper, and periodically drummed.

The C-340 operation was also capable of re-melting the U derbies and casting specific shapes. Operations were conducted in a furnace with a controlled atmosphere. Graphite crucibles were used to receive the molten U. The primary hazard associated with these operations was cleaning the crucibles between pours. Over time, oxides of U and beta-emitting U decay products would impregnate the crucible. Since crucibles were cleaned by hand, operators would have received radiation dose to their hands, arms, and fingers (Phase II Investigation). No dosimetry was worn by operators that would have measured these extremity exposures.

#### **2.2.4 Recovery Operations**

During PGDP's operational history, U has been recovered from waste streams and recycled through the enrichment process to minimize loss of this valuable material. Np and  $^{99}\text{Tc}$  were also recovered during early PGDP operations to meet high demands for these materials. Recovery operations reduced the releases of U, Np, and  $^{99}\text{Tc}$  to the environment but produced high concentrations of radioactive materials in PGDP processes that potentially posed significant occupational hazards to workers.

The source of Np and  $^{99}\text{Tc}$  at PGDP was feed material from U recovered from spent reactor fuel at the Hanford and Savannah River sites. The AEC understood that FP and TRUs could present health problems to gaseous diffusion workers and set limits on the amount that could be present in feed materials (Phase II Investigation). The chemical separation processes at Hanford and Savannah River removed most, but not all, of the TRUs and FP.

#### **Uranium Recovery**

U recovery facilities in C-400 were used to chemically separate and recover U from a variety of waste materials. Sources of feed material for this process included: fluorination tower ash, sintered metal filters, decontamination solutions,  $\text{UF}_6$  scrubber solutions, particulates from ventilation filters and vacuum cleaners, laboratory wastes, and materials from spills. Before the mid-1970s, a complex U recovery process in Building C-400 separated U from waste and scrap materials, concentrated it, and converted it to an oxide. The process included the following steps: dissolution of feed materials, filtration, solvent extraction in pulse columns, concentration by evaporation, and denitration to an oxide.

The U recovery system was not leak-tight, and leaks were common. Steps were taken to control operators' exposure to process materials. Routine surveys were conducted to monitor the concentration of radioactivity on surfaces and in the air in C-400, and the health physics staff recommended changes in work practices based on the results of these surveys. U recovery system operators were provided coveralls. Rubber gloves and respirators were available, but their use

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was not strictly enforced; they were generally worn at the discretion of the operators. The aqueous raffinate waste from solvent extraction columns that contained  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$ ,  $^{234}\text{Th}$ , and  $^{99}\text{Tc}$  was discharged to the environment.

In the mid-1970s, the solvent extraction process for U recovery was replaced with a simpler precipitation and filtration process. Steps in this new process included: dissolution of feed materials in nitric acid, addition of lime to precipitate U, and recovery of precipitated U as a filter cake.

The filtrate, containing low concentrations of radionuclides, was discharged to the environment. Sludges and filter cake were buried on site if U concentrations were low or sent to FMPC if concentrations were high enough to warrant further recovery.

### **Neptunium Recovery**

Soon after Np was identified at Paducah in 1957, the AEC placed a high emphasis on its recovery. A Np recovery process was developed at ORNL, and began operation at PGDP in November 1958 in Building C-400. The process used a solvent extraction and evaporation method to recover and concentrate Np from receiver ash and cylinder heels wash solution.

Receiver ash and solids that settled from cylinder wash water were dissolved in a nitric acid solution. Solids suspended in this solution were removed by filtration and discarded as solid waste. The filtrate was processed through solvent exchange pulse columns to separate U, Th, and Np. (These columns were originally located in Building C-710, Room 32, and may have been moved to C-400 sometime after July 1959.) Raffinate from these columns was dumped to the building drain if it contained U and Np concentrations less than 500ppm and 2.0 mg/L, respectively. U and Th were recovered for future use. The Np solution was concentrated to about 20 to 25 g/L by evaporation. The concentrate was sent to a laboratory in Building 710 for additional separation and concentration in ion exchange columns. The final product was siphoned into glass carboys on the loading dock at C-710 and shipped to ORNL.

The highest concentrations of Np at PGDP were associated with Np recovery processes that operated intermittently from 1958 until the late 1970s. These processes separated and concentrated Np from receiver ash, cylinder wash water, and  $\text{MgF}_2$  pellets used in  $^{99}\text{Tc}$  traps. One liter of Np recovery product contained about one curie of radioactivity (Phase II Investigation).

The relatively high hazards associated with Np were understood at Paducah as early as 1959, and special practices for handling Np solutions and Np-contaminated equipment were recommended. Recommendations included: using non-breakable containers; maintaining tight systems; keeping lids on containers; preventing bubbling, frothing, or spraying of solutions; using rubber gloves; washing the gloves before using them in other areas; using respirators (or assault masks) for welding or burning; and performing alpha surveys of all equipment removed from Np processing areas.

Estimates show that 4.289 kg of Np were recovered using the above process (3.215 kg from heel washings and 1.074 kg from ash) (Phase II Investigation). This process was discontinued in October 1961, after  $\text{MgF}_2$  traps were determined to be a more productive method of recovery. The recovered Np was shipped from the site. The Np recovery system was removed from the Plant in the late 1970s.

The processing of solutions containing Np through the solvent extraction and ion exchange system produced raffinate and wash solutions with some Np remaining. Solutions with Np concentrations greater than 2 mg/L were either reprocessed or stored. Eighteen drums of waste from the Np recovery program remain stored on site today. Solutions with a Np content less than 2 mg/L were discharged to the environment using building drains. Estimates indicate that approximately 200 grams were discharged as a result of recovery operations (Phase II Investigation).

A second Np recovery process was used briefly after 1961 to recover Np from  $\text{MgF}_2$  pellets that had been removed from  $^{99}\text{Tc}$  traps in the feed plant and cascades. Although the traps were originally installed to adsorb  $^{99}\text{Tc}$ , they were also quite effective in adsorbing Np. The pellets were vacuumed from traps in the feed plant and cascades and transported to Building C-400, where a chemical stripping process removed Np. Approximately 33 grams of Np were recovered by this method before recovery operations were terminated at the site in the mid-1960s (Phase II Investigation).

### **Technetium Recovery**

$^{99}\text{Tc}$  is a fission product that was received at Paducah in recycled feed from Hanford and Savannah River Sites.  $^{99}\text{Tc}$  passed through the Paducah cascade as a volatile compound of fluorine, depositing on internal surfaces of the cascade and contaminating the enriched U product. The AEC did not specify a limit for  $^{99}\text{Tc}$  in  $\text{UF}_6$  feed but controlled the concentration of  $^{99}\text{Tc}$  indirectly to about 10ppm by limiting gross beta from FP (Phase II Investigation).

A demand for  $^{99}\text{Tc}$  in the early 1960s prompted Paducah to begin a process to recover 25 kg of this material from various effluent streams. In 1960, a process was begun to recover  $^{99}\text{Tc}$  from  $\text{UF}_6$  cylinder wash water and from the raffinate generated during Np recovery. Process steps included precipitation and removal of U from these solutions by adding sodium hydroxide. The aqueous superannuate was processed through an ion exchange column and elutriated with nitric acid to produce a concentrated solution of  $^{99}\text{Tc}$  that was shipped to ORNL. Although  $^{99}\text{Tc}$  was not a significant radiological hazard during most PGDP operation and maintenance activities, this concentrated form presented a more significant hazard.

$^{99}\text{Tc}$  traps were installed in the feed plant and in the cascades in 1961 and 1963, respectively, to reduce contamination of the enriched U product. A small amount of  $^{99}\text{Tc}$  was recovered from these traps in the early 1960s.  $^{99}\text{Tc}$  was leached from the pellets in a dissolver in C-400 and potassium hydroxide was added to precipitate the U. The solution was then filtered and processed in the same manner discussed above.

In the mid-1970s, a process was developed and implemented at PGDP to remove  $^{99}\text{Tc}$  from aqueous waste streams for the purpose of environmental protection.  $^{99}\text{Tc}$  in superannuates following U precipitation was removed as an insoluble solid through the use of iron sulfate as a flocculating agent and collected as waste sludge.

### **2.3 Activity Summaries (concentrating processes and other site-specific issues related to processing and plants)**

Several plant activities resulted in the concentration of TRU and fission product materials to levels potentially more radiologically significant than the U materials handled at the plant. The processes of concern for the concentration of the TRU and fission product materials are summarized in Table 2.3-1, Transuranic and Fission Product Concentrating Processes.

<b>Table 2.3-1, Transuranic and Fission Product Concentrating Processes</b>	
Np in Ash (K/ETO-30)	The C-410/C-420 feed plant operated July 1953 through June 1964 and July 1968 through June 1977. It is estimated that approximately 25% (20%) of the Np in the feed material remained in the ash. Some of the ash was processed through the recovery process.
Np in Cylinder Heels	UF <sub>6</sub> containing TRUs and fission product materials was transferred from the feed plant to UF <sub>6</sub> cylinders used to feed the cascade. 75% of the material initially present was transferred to the cylinders. Approximately 25% was fed to the cascade. The Np remaining in the cylinders was eventually processed through the C-400 cylinder washing facility. Some of this material would have remained in the cylinder as material bonded or plated to the cylinder internal surface. The majority of the material would have been processed through recovery operations or discharged as C-400 decontamination wastes or effluents.
Np in Cascade (K/ETO-30)	Studies have indicated that 6.3 kg (4.6 kg) of Np have been fed to the cascade. No Np has been detected in the tails withdrawal and only negligible quantities have been detected in product material. It is estimated that 4.7 kg (1.3 kg) of Np were removed during cascade improvement programs resulting in 1.6 kg (1.02 kg) remaining in the cascade. The equipment removed, as part of the cascade improvement programs would have gone through the recovery operations or discharged as C-400 decontamination wastes or effluents.
Np Recovery Operations	Ash and cylinder washings were processed through the C-400 Np recovery process. The process used aqueous chemistry and ion exchange methods to recover 3.215 kg Np from the cylinder wash stream and 1.074 kg Np from the ash stream. The recovered materials were shipped offsite to the Hanford site.
<sup>99</sup> Tc in the Cascade and Product Cylinders	Approximately 616 kg of the <sup>99</sup> Tc received at PGDP was available for feed to the cascade. Of this amount approximately 560 kg would be fed from the cylinder to the cascade. This material would rapidly migrate up the cascade and either be withdrawn with the UF <sub>6</sub> product or vented by the purge cascade.
<sup>99</sup> Tc Recovery Operations (KY/L- 1239)	<sup>99</sup> Tc was processed in recovery operations during the early 1960s. Approximately 25 kg were recovered and shipped to ORNL.
Pu in Ash	Approximately 300 grams of Pu were introduced to the PGDP feed plant from the recycled reactor returns from Hanford and Savannah River. It is estimated that only 0.3 grams of the Pu was introduced to the cascade through feed materials and the remainder concentrated in the ash byproducts for the feed production plant. Any Pu exposure would be mostly attributable to personnel working in the dust environment of the ash collector and filter.
In Cascade	Most of the 0.3 g of Pu would have been removed in CIP/CUP.



## **2.4 Activities where workers were likely to be in contact with recycled uranium through direct physical contact or through airborne dust**

### **Major Component Maintenance**

Maintenance on major components in the cascade (compressors, converters, and process block valves) presented some of the most significant opportunities for exposure of maintenance personnel. Work on these components required that they be removed from the system, cleaned, rebuilt or repaired, and then reinstalled. In order to remove these components, process operators isolated and bypassed the cascade cell containing the component, reduced the UF<sub>6</sub> within the cell to less than 10ppm equivalent at atmospheric pressure (a UF<sub>6</sub> negative), and then purged the cell to minimize HF and UF<sub>6</sub> exposure of workers involved in opening, maintaining, or modifying cell components. Once a satisfactory UF<sub>6</sub> negative and HF purge was accomplished and the pressure of the isolated cell was raised to atmospheric pressure with dry air, the isolated cell was turned over to process maintenance for cell opening and disassembly.

Workers opening a cell and dismantling cell components could be exposed to UF<sub>6</sub>, HF, UO<sub>2</sub>F<sub>2</sub>, and to a lesser extent, TRUs and FP, such as <sup>99</sup>Tc. Maintenance personnel would initially make a small hole or cut in the process gas piping to confirm that cell pressure was at approximately atmospheric pressure. To prevent the potential spread of radioactive contamination, the same maintenance procedure required all openings into components to be covered as soon as practicable after removal from the process gas piping.

Compressors were transported from the process buildings to Buildings C-720 and C-400 for "000" and "00" sizes, respectively ("000" and "00" are size designations, with "000" being larger). The compressors were then disassembled into major components within pits, the parts transported to Building C-400 for spray washing to remove U deposits, the rotor and stator relocated as required for deblading within C-400 and C-410, respectively. All of the reusable washed parts were returned to their respective maintenance buildings for modification, refurbishment, degreasing, and reassembly. Once reassembled, the compressor openings were covered for transportation to storage or reinstallation. Converters were transported from the process buildings to Building C-409 for decontamination. The barriers were then taken to Building C-400 for washing, disassembly, and scrap recovery. Following washing in C-400, the converters were modified, refurbished, and reassembled in Building C-720. Prior to removal from the system, block valves were slightly opened (where possible), inspected, cut out of the system, lifted free of process piping, decontaminated, covers installed, and shipped to C-400 for preliminary disassembly and decontamination to the limits allowed in C-720. Once decontaminated, the valve was again covered and transported to C-720 for final repair and reassembly, and staged in the process building for reinstallation.

During the 1970s and early 1980s, AEC/ERDA/DOE and Union Carbide undertook the most extensive of several campaigns to improve PGDP technology and exchange or replace aging equipment (CIP/CUP). All of the industrial, radiological, and chemical hazards discussed for normal compressor and converter maintenance were present, with the additional challenge of a demanding, manpower-intensive schedule for completing each task. Dedicated cell change-out teams were established to remove and replace cell components almost continuously. Tools for cell change-out were pre-positioned. Cell housings were opened as operators worked to establish a UF<sub>6</sub> negative. Modified and refurbished compressors and converters were pre-staged in the

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process buildings with proper orientation, ready for emplacement once the cells were cleaned out and new saddles and support systems installed. Original cell components were disassembled, cleaned, modified, refurbished, reassembled, conditioned, and pre-positioned for another cell change-out, even as the original cell was being repopulated. Operators were prepared to perform leak checks, pre-operational tests, and cell startup as soon as maintenance approved the release of the various permits establishing their safety envelope. Many workers were hired to support CIP/CUP, but reportedly they did not get the same level of training as older workers; they were told to rely on more experienced workers while learning their jobs, principally through on-the-job training.

Practices to minimize personnel exposure to airborne radioactivity in the shops evolved over time. In 1959, recommendations were made for additional dust control measures to minimize the potential for exposure. These included use of continuous water mist spray during removal of the compressor stack and collection of the resulting wash water, wearing air respirators in the C-720 pit area until lower air counts were obtained, disassembling compressors to three main components and removing them to C-400 for spray decontamination, wetting down compressor spool piece bolts prior to air tool removal, decontaminating compressor mating pipe flanges in the original cell area prior to grinding, and removing slag. Despite ongoing work to improve the local area exhaust in the C-720 converter shop, health physics also recommended thorough wetting of disassembly work while workers continued to wear respirators. In 1962, at least one sample of dust from C-400 compressor disassembly showed 90 percent of its radioactivity from TRUs and FP (Phase II Investigation). Although dust was removed by vacuuming, the rotor was not wetted to control dust as required. Respirator use was noted to be "as required." (Phase II Investigation)

As CIP/CUP progressed in the late 1970s, so did the degree of sophistication of the health physics survey reports. Levels of U, Np, Pu, Th, <sup>99</sup>Tc, and U daughter products were routinely reported and discussed, with accompanying recommendations. Contamination surveys just outside the compressor pit area prompted a call for better housekeeping practices. Continuous air samples near the pit and adjacent machine shop indicated no significant spread of airborne radioactivity to the surrounding area. During obviously dirty job steps, respirators were reportedly used; however, respirator use was still observed to be lax during many short-duration tasks.

In 1976, the Health Physics and Industrial Hygiene Department concluded that methods established to that date for control of personnel exposure during compressor maintenance were adequate, but emphasized the importance of maintaining these practices. The practices included respiratory protection using one-quarter or one-half respirators for protection against radioactive aerosols or radionuclide filter cartridges for certain specified jobs; vacuuming loose material, dust deposits, and spilled material; wetting down compressor stacks with water before placing them in the disassembly stand; collecting wash water for delivery to C-400; and decontaminating compressor parts in C-400 after stack disassembly. Despite these recommendations, problems with respirator use continued to be reported (though less often). The Health Physics and Industrial Hygiene Department reminded management of the importance of respirator use while disassembling converters in C-409, particularly in light of the high levels of TRUs detected in solid deposits within the converters. Concern was again expressed over the lack of adequate local air exhaust in the C-409 converter shop areas where dust-producing activities were performed.

In 1977, continued attempts to establish adequate local area exhaust and stop the use of the air mover in the compressor pits were at first unsuccessful. The Health Physics and Industrial Hygiene Department recommended continued efforts to stop dust generation at the source as an ALARA principle. Further, the Health Physics and Industrial Hygiene Department recommended immediate action to provide adequate exhaust ventilation, supported in part by breathing zone air

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samples exceeding Plant guidelines for U, Np, and Th by factors of 40, 22, and 15, respectively (Phase II Investigation). The Health Physics and Industrial Hygiene Department also recommended continuing use of the vacuum collector system for loose deposits, keeping compressor components wet during use of pneumatic tools, and providing local air exhaust to all disassembly steps where practical. The Health Physics and Industrial Hygiene Department noted that additional local area exhaust was being designed and would be installed as soon as possible in C-409 to support converter disassembly work. The Health Physics and Industrial Hygiene Department also recommended the use of water sprays in C-400 to control dust during barrier disassembly.

In 1978, The Health Physics and Industrial Hygiene Department commended the shops for use of low-speed, high-torque wrenches and ventilation upgrading by extending the vacuum system to an adapter on the pneumatic wrenches. Collecting the dust at the source of generation was noted to decrease concentrations of U by 98 percent and Np by 91 percent (Phase II Investigation). However, this fix did not survive the rigors of compressor maintenance work and was later abandoned.

Health physics surveys of the C-720-C converter shop in 1980 for the CIP/CUP indicated that Plant guides for airborne alpha activity were exceeded for U by a factor of 1680, <sup>237</sup>Np by a factor of 2121, <sup>239</sup>Pu by a factor of 2483 and <sup>230</sup>Th by a factor of 55. Even using conservative protection factors for the respirators used, these exposure levels were significant. (Phase II Investigation)

The levels of airborne contaminants resulting from these maintenance activities, supervisors' failure to enforce proper use of respirators, and employee's failure to wear respirators when required may have contributed to the high proportion of personnel who were on restriction for elevated levels of U in their urine. For example, a sample of exposure records from the first half of 1978 shows that 20 of 29 urine samples exceeding the PGDP investigation level were from individuals involved in CIP/CUP activities (Phase II Investigation).

### **Cylinder Cleaning**

With repeated reuse, UF<sub>6</sub> cylinders collected deposits that did not completely volatilize in the autoclave. Periodically these deposits, called "cylinder heels," had to be dissolved and removed, and the cylinder was then cleaned, refurbished as necessary, re-inspected, hydrostatically tested, and weighed for subsequent use. Cylinder heels were composed of corrosion products, U salts and oxides, and TRU and U daughter product compounds. With regard to the contaminants of the process gas, some of the Np and much of the <sup>99</sup>Tc was volatilized to the cascade, while most of the Pu remained behind in the cylinder heels, creating a significant radiological hazard. Cylinder cleaning was performed at Building C-400, where the heels were dissolved and the rinse water was collected in a large pan. Cylinder rinse water was used as the principal source for Np and <sup>99</sup>Tc recovery in the late 1950s and early 1960s. Otherwise, liquid effluents were pumped to the tank farm for feed into one of the digesters, while workers shoveled sludge, which collected in the pan, into containers for further processing or disposal. Sludge reportedly was shoveled approximately once a month; workers were limited to 15-minute exposures, and it usually took four workers to complete the task. The Health Physics and Industrial Hygiene Department closely monitored worker activities. (Phase II Investigation)

Two documented beta overexposures occurred at the C-400 cylinder wash facility in the first quarter of 1968 (Phase II Investigation). The estimated exposures were 24 and 36 rem, whereas the quarterly limit for skin of the whole body was 10 rem. The two workers were standing in a metal tray used for collecting cylinder rinse water that was emitting several hundred rads of beta

radiation. The evaluation for the cylinder wash overexposure incident failed to completely evaluate this event and determine extremity dose. (Phase II Investigation)

### **Cylinder Valve Replacement**

Each UF<sub>6</sub> cylinder is equipped with a manual cylinder valve. Occasionally, these valves were identified as defective and would be replaced. According to procedures that existed in the 1970s, any UF<sub>6</sub> cylinder was required to cool at least five days before its valve was replaced (Phase II Investigation). Cylinders known to be above atmospheric pressure after the minimum cooling period would be cold-burped and further cooled, if necessary, with cold water. If pressure above atmospheric could not be relieved, the cylinder would be turned over to Chemical Operations in C-400 for special handling, which involved dedicated tanks used to further cool the cylinders to promote UF<sub>6</sub> solidification and pressure reduction.

Cylinder valves were normally replaced in C-310, C-315, or the tails storage area. Valve replacement during the 1960s also occurred in the vicinity of C-400, after icing down cylinders. Until the mid-1970s, defective UF<sub>6</sub> cylinder valves were routinely replaced "on the fly" with the mechanic standing upwind and any escaping smoke going the other way (Phase II Investigation). The applicable maintenance procedure in the 1970s and 1980s required respiratory protection to be worn; however, even though gas masks were available, they were not always utilized until a release of HF ("blow-out") occurred. The defective valve was slightly unscrewed to confirm that air would be drawn into the cylinder. Once a vacuum was confirmed, the valve was quickly removed and the replacement valve installed. If positive pressure was evident on the first attempt to change the valve, the original valve would be retightened and another attempt scheduled in not less than 24 hours. If positive pressure was still noted on the second attempt, the valve would again be retightened and the cylinder would be turned over to Chemical Operations in C-400 for special handling. Once the valve was successfully replaced with the proper torque and thread engagement, the defective valve was decontaminated and appropriately dispositioned. The new valve and cylinder combination was then inspected and pressure tested to confirm a successful repair.

In the event of a major UF<sub>6</sub> release from an open or broken cylinder valve, procedures in the 1970s provided guidance that personnel should be immediately evacuated from the area of the release, emergency assistance summoned, and available emergency ventilation maximized. Caution was provided to stay upwind of the release; that personnel required to enter the release area must wear Gra-Lite, Acid Master, or impermeable suits with self-contained air masks; that exposed personnel should report to the dispensary as soon as possible; and that all water in the area should be considered contaminated with HF and neutralized with soda ash. The emergency squad was expected to apply water to the cylinder to promote cooling and knock down the UF<sub>6</sub> cloud, stop the leak with a wooden plug or tape if the valve could not be shut, and (if that didn't work) cover the cylinder with a prefabricated box from C-310, filling the box with dry ice and covering with a tarpaulin. Once the cylinder could be cooled to the point of drawing a vacuum, the defective valve was removed and a replacement valve installed.

A Three-Plant UF<sub>6</sub> Cylinder Handling Committee convened in the mid-1970s and made a number of recommendations that affected PGDP cylinder valve replacement activities. Among the recommendations implemented by 1986 were the sole use of new valves for valve repair or replacement, modification of procedures for valve replacement to drop reference to freeze-down tanks at C-400 (although the tanks existed, onsite supplies of dry ice were insufficient for emergency or contingency use), and revision of site procedures to address the use of updated

emergency release securing equipment and new studies indicating that water should not be used on liquid UF<sub>6</sub> releases.

### Filter Bag Replacement

Filter bag houses existed in several buildings for both ventilation and dust collection. Replacing the bags in these systems was very dusty and dirty work. Workers were periodically directed to replace the filter bags because of excessive dust loading. Filter bags needed to be changed once or twice a month, but the same individuals did not always get the assignment due to shift work. In the 1950s, workers reportedly secured the evacuation jet, donned army assault masks and a company-provided coat over their company-provided coveralls, draped towels over their heads and around their necks, taped their sleeves up, opened the enclosure, released the hose clamps in sequence, and carefully put the dusty bags in large barrels. Operators then vacuumed the remaining dust from the enclosure, and maintenance installed new filter bags, closed the enclosure, and started the evacuation jets again. In the early 1960s, concern about radiological exposure resulted in reducing workers' times in the area to no more than 15 minutes, significantly less than previously allowed. (Phase II Investigation)

Some workers in C-340 and C-420 changed filter bags without respirators or anti-contamination clothing. Sometimes small paper masks were used, even though they came out covered in green dust. During occasional periods between 1968 and 1977 the C-410 or C-420 bag houses were bypassed straight to the atmosphere whenever they got plugged or needed changing (Phase II Investigation). Hazards to workers included airborne UF<sub>4</sub>, uranium oxides, process dust, and alpha and beta contamination. Workers wore dosimetry devices and were subject to monthly bioassays. Respirators occasionally became plugged and were sometimes not used (Phase II Investigation). When filter bag replacement activities were evaluated by health physics, they were found to be dusty and often presenting the potential for elevated external exposure.

### Assessment of Potential Exposure

Assessments of activities that are assumed to have most likely involved potential worker exposure to RU constituents are described in Table 2.4-1. Worksheets developed to support these assessments are located in Appendix F. Factors used to assess Occupational Potential Exposure (OPE) and methods used to calculate constituent dose (Prioritization of Uranium Flows, Appendix A of Appendix F) as a percent of the dose from a specific uranium stream are also included in Appendix F.

**Table 2.4-1, Assessment of Activities at Paducah where workers were most likely to contact Recycled Uranium**

	Location	Activity	Time Frame	Potential Constituents of Interest- Maximum concentrations of constituents of interest per Activity Worksheet:	Occupational Potential Exposure Rating
1.	C-333 C-337 C-410	<b>Cascade Operations-</b> Operators could have been exposed to gaseous UF <sub>6</sub> when connecting or disconnecting feed cylinders containing recycled uranium.	FY-1953-1964 FY-1969-1970 FY-1972-1976	450 ppb Np .09 ppb Pu 23,000 Tc <sup>99</sup> (UF <sub>6</sub> )	<b>2 Moderate</b>

	Location	Activity	Time Frame	Potential Constituents of Interest- Maximum concentrations of constituents of interest per Activity Worksheet:	Occupational Potential Exposure Rating
2.	C-331 C-333 C-335 C-337 C-400 C-409 C-720	<b>Cascade Maintenance-</b> Maintenance workers could have been exposed to constituents of interest when working on or removing the interior surfaces of cascade equipment near recycled uranium feed points (Cascade Dust)	FY-1954-1961 FY-1973-1981 (CIP/CUP)	2740 ppb Pu 3,220,000 ppb Np XX Tc <sup>99</sup> (Cascade Dust)	<b>3 High</b>
3.	C-410 C-420	<b>Feed Plant Operations, RU to UF<sub>6</sub>-</b> UO <sub>3</sub> → UO <sub>2</sub> UF <sub>4</sub> UF (handling, drumming, bag changing etc.) (Tower Ash)	FY- 1953-1964 FY-1969-1977 FY-1982-1983 (Shipped)	25,602 ppb Np 8,000 ppb Pu 4,600 ppb Tc <sup>99</sup> (Tower Ash)	<b>3 High</b>
4.	C-400 C-710	<b>Uranium/Neptunium Recovery-</b> Workers salvage uranium from cleaning solutions and waste products. (Cylinder wash, Ash, dust). Aqueous Solutions (Solutions containing >2mg/L Np were reprocessed, <2mg/L discarded.)	FY-1952-1990 FY-1956-1976	25,000,000ppb Np 100,000ppb Pu XX ppb Tc <sup>99</sup> (Solutions)	<b>2 Moderate</b>
5.	C-340	<b>Production and Handling of UF<sub>4</sub></b> <b>Produced from Tails-</b> UF <sub>6</sub> , UF <sub>6</sub> → UF <sub>4</sub>	FY-1957-1962 FY1967-1977	62.6ppb Np .063ppb Pu 49ppb Tc <sup>99</sup> (UF <sub>4</sub> )	<b>De Minimis</b>
6.	C-405	<b>Handling Ash from Contaminated Items-</b> Incinerator (filter bags, etc.)	FY-1953-1990	3,512ppb Np 56ppb Pu 921ppb Tc <sup>99</sup> (Ash)	<b>3 High</b>
7.	C-315 C-340	<b>Connecting and Disconnecting UF<sub>6</sub></b> <b>Tails Cylinders and Handling Tails UF<sub>6</sub></b>	FY-1953-1999	5ppb Np 0.1 ppb Pu 20,000ppb Tc <sup>99</sup> (UF <sub>6</sub> )	<b>De Minimis</b>
8.	C-310	<b>Connecting and Disconnecting UF<sub>6</sub></b> <b>Product Cylinders and Handling Product UF<sub>6</sub></b>	FY-1953-1999	27ppb Np .06ppb Pu 20,000ppb Tc (UF <sub>6</sub> )	<b>De Minimis</b>
9.	C-410 C-310	<b>Changing/Cleaning Magnesium Fluoride (MgF<sub>2</sub>) Traps</b>	FY-1964-1966	1900ppb Np .01ppb Pu 2,266,000ppb Tc (MgF <sub>2</sub> )	<b>2 Moderate</b>

	Location	Activity	Time Frame	Potential Constituents of Interest- Maximum concentrations of constituents of interest per Activity Worksheet:	Occupational Potential Exposure Rating
10.	C-340	<b>U-Metal Production from metal production, sawing, and metal handling</b>	FY-1957-1962 FY-1967-1977	2.7ppb Np .0085ppb Pu 10.6ppb Tc (U-Metal)	<b>De Minimis</b>
11.	C-340	<b>Handling MgF<sub>2</sub> in U-Metal Manufacturing-</b> cleaning, roasting, liner preparation, knock out and drumming.	FY-1957-1962 FY-1968-1977	2,400ppb Np 1.2ppb Pu 1,880ppb Tc (MgF <sub>2</sub> )	<b>3 High</b>

\* xx = No Data Available

## 2.5 Workforce Monitoring and Radiological Protection Practices and Records

### Programs and Controls

Health and safety programs at PGDP were established at the commencement of Plant operations and continue to the present day. Health physics, industrial hygiene, and medical functions were integrated in the Health Physics and Industrial Hygiene Department for the first three decades of Plant history, and under the direction of the Plant Medical Director, this integrated several safety disciplines with a focus on worker health. From the commencement of operations until the Tiger Team evaluation in 1990, both health physics and industrial hygiene were minimally staffed, especially in comparison with the number of safety professionals that would be required today for the types of hazards and work activities present. The Health Physics Section from the commencement of operations until 1990 ranged in size from as few as two to six employees. The Industrial Hygiene Section typically consisted of one or two industrial hygienists and a technician. Furthermore, in the early decades, health and safety professionals had limited authority and resources to ensure that line management would implement recommended hazard controls. The primary responsibility for protecting personnel against hazards associated with radioactive materials was placed on line supervision to the same extent that they were responsible for operation and production.

During the first three decades, the Health Physics and Industrial Hygiene Department provided workers and line management with the following basic programs and services:

- Monitoring exposures to determine the effectiveness of the health physics program;
- Auditing and maintaining records of exposures (radiological, noise, chemicals) and radiation data collected throughout the Plant;
- Furnishing line supervisors with advice, information, and training aid on chemical, radiological, or U toxicity health hazards;
- Assisting in investigations of personnel exposures;
- Providing film badge services;
- Maintaining the bioassay and respiratory protection program for both chemical and radiological exposures;

- Performing chemical and radiological environmental monitoring for the Plant;
- Recommending radiological and chemical Plant guidelines for controlling exposures; and
- Conducting air sampling for airborne chemicals and radioactive material.

As early as the 1950s, PGDP set forth in policy and Plant procedures the expectations for the protection of personnel from the hazards inherent in handling radioactive materials (Phase II Investigation). The policy states that "every effort is made to prevent personnel exposure from exceeding the Radiation Protection Guideline established by the Federal Radiation Council, the provisions of the AEC Manual Chapters" (subsequently ERDA and DOE), "or those established by the National Committee on Radiation Protection and Measurements; the maintenance of radiation doses as far below these standards as is practical is also encouraged." (Phase II Investigation).

### **External Exposure Monitoring Programs**

Both the Health Physics and Industrial Hygiene Department and line management monitored external radiation exposures at PGDP from the 1950s to 1990. The Health Physics and Industrial Hygiene Department was responsible for performing beta-gamma radiation monitoring of the general work areas, equipment surfaces, material shipments, and personnel on a routine and spot basis and reporting findings to appropriate supervision with any necessary recommendations. The responsibility for performing routine radiation detection surveys lay with the line division concerned with the work being performed. Each division was responsible for identifying equipment having significant radiation exposure potential and establishing work time limits.

Personnel exposures were primarily monitored by the use of film badges. After July 1, 1960, film badges were assigned to all employees, and were supplied to all individuals who visited the Plant from other locations and who might have been exposed to as much as one-tenth the RPG. Before July 1960, only selected workers were included in the film badge service based on their work activities. For example, in 1956 and 1958, there were 350 and 450 employees in the film badge service, respectively. Before 1960, the basic film badge use period appeared to be one week; in the early 1960s, the period was extended to one calendar quarter. However, for employees whose work involved significant exposure and who might have exceeded the quarterly RPG, badges were read and exchanged monthly. Employees on the monthly exchange cycle were primarily involved in chemical processing, maintenance of chemical processing facilities, and U metal production (Phase II Investigation).

In general, the low specific activity and the self-shielding properties of U limited dose rates at PGDP. However, certain operations were known to result in concentrations of material having higher specific activity and having created conditions that, if undetected, could result in exposures above permissible limits. Routine whole-body beta exposures over PGDP investigation levels existed primarily at areas where U daughter products and TRUs tended to concentrate, including the feed plant fluorination towers, converter disassembly areas in C-400 and C-720, the cylinder wash facility in C-400, the C-340 metals plant, and the Np and U recovery process raffinates. Exposures to external radiation were managed to assure that no one went above their lifetime limit (5N-18 rem, where N is a worker's age in years) (Phase II Investigation). It was common to rotate workers through areas of high external exposure concern, such as the ash receiver area, to administratively control individual exposures.



In the early 1950s, extremity monitoring was not required because it was felt that these doses were not likely to exceed 2.5 times the whole-body exposure. Whole-body exposures to operators and the dose rates in the ash receiver area were large enough that they could exceed 10 percent of the extremity limit and, therefore, would necessitate extremity monitoring (Phase II Investigation). Shell and crucible cleaning operations in the metals plant required time-consuming wire brushing. In this activity, an individual would reach into a mold containing oxides rich in U daughter products (primarily beta emitters) and physically clean off the materials from the walls and bottom. The individual's film badge, worn on the torso, would typically be shielded from the majority of the beta activity by the crucible itself. However, since the whole body exposures to operators and the dose rates from these shells and crucibles are large enough that they could exceed 10 percent of the extremity limit; this practice would also have necessitated extremity monitoring (Phase II Investigation). However, Health Physics and Industrial Hygiene Department summary reports provided no extremity monitoring data. Two documented, known beta overexposures (skin of the whole body quarterly limit) occurred at the C-400 cylinder wash facility during the first quarter of 1968. Investigation of this event did not address or determine extremity dose (Phase II Investigation).

#### **Bioassay - Urinalysis Programs**

Individual employees were required to submit urine specimens for U analysis at a frequency thought to be commensurate with exposure potential, as well as for periodic physicals. Additionally, special urinalyses were scheduled for those working on special jobs, or when some special investigative information was required. The frequency of routine urine samples, for U, varied from a maximum frequency of four weeks for all personnel working in chemical operations and metal production (primarily C-310, C-315, C-340, C-400 and C-410) to a minimum frequency of 12 months for those working in locations deemed to have little likelihood of exposure (Phase II Investigation). The Health Physics and Industrial Hygiene Department routinely issued a master schedule to line management showing when specific samples should be taken from certain groups of employees. This schedule typically covered three calendar months. Action points for U levels in urine were established, setting forth recall-sample frequencies, supervisor notification, and investigation reports. These action points ranged in levels from just above detection capability to greater than RPGs (Phase II Investigation). The actions that were taken were commensurate with the result, typically ranging from requiring recall samples, workplace investigation, workplace restriction, estimate of body burden, and internal dose and/or confirmatory in vivo radiation monitoring (e.g., lung counting).

Employees who were administratively removed from work because of exposures were reassigned to areas with less potential for intake, although typically still in areas where U work was conducted. The urinary U excretion rates were followed for these individuals until the urinalysis results were understood from a solubility standpoint or until rates decreased to baseline values; the personnel then returned to their regular work activity. Biological retention times for these types of exposures are closely related to the solubility class of the compound. Although the health physics group actively tried to gain insight into solubility class and particle size, much of this information was not well understood during the early 1950s and 1960s. (Phase II Investigation)

#### **Bioassay - In Vivo Radiation Monitoring (Information from Phase II Investigation)**

In vivo radiation monitoring via lung counting for PGDP workers was conducted initially at fixed facilities at FMPC and Y-12 in Oak Ridge. Later, PGDP conducted a vivo radiation monitoring using a mobile system from Oak Ridge. Data indicated monitoring for U, Np, and <sup>99</sup>Tc, and generally indicated no significant accumulation of radioactive material in the lungs in excess of

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RPGs. However, the PGDP quarterly report for July-Industrial Hygiene summary, indicated that a PGDP maintenance mechanic who had been checked in the Y-in effect at this time. Radiation Protection Criteria and Standards, Their Basis and Uses at AEC micrograms per day may be considered indicative of a significant internal body deposition of normal U." early in vivo radiation monitoring conducted at, or for, PGDP.

In vivo monitoring was often conducted following discovery of elevated levels of material in air or urine of 1979 for the C-time period indicated that several personnel working at C-ng converter bundle salvage had elevated lung deposits of U. Pu was detected in some air samples in significant concentrations

990, the PGDP used a network of stationary air samplers at various production and non production areas throughout the Plant. Portable and breathing zone samplers supplemented this network. Much of the data indicated frequent air sampling results in excess PGDP RCG levels. Health Physics and Industrial Hygiene monthly summary reports between 1955 and 1968 indicated that it was common to have air samples collected by both stationary and ally were related to a process upset, equipment failure, or maintenance activity.

Several air samples that were collected during the first quarter of 1962 in conjunction with The total alpha activity from Np in sample results ranged from non detectable to greater than 90 percent. A review of these evolutions also showed examples of airborne contamination ranging from non detectable to more than 100 times the PGDP MPC for Np. Health Physics and Industrial Hygiene

as continuing to present a difficult exposure control problem. Health Physics and Industrial Hygiene Department surveys of CUP w-720 C Converter Shop in 1980 indicated that Plant guides for airborne alpha activity were exceeded for U by a factor of 1,680, Np by a factor of 2,121, Pu by a factor of 2,483, and <sup>230</sup> factor for respiratory protection afforded by a respirator ("conservatively is 90% effective"), -tenth as great would be deemed significant. The specific operations identified as acetylene torch to cut through jackscrews from inside the converter and use of compressed air blow through testing. Both were subsequently abandoned.

tends to indicate that contamination control practices were lax at Paducah from the beginning of operations until the mid 1980's. In the mid-NRC and DOE promulgated more stringent contamination control and radiological release criteria. While were desirable, these practices were neither rigorously enforced nor mandatory during the early years of PGDP operations. Radioactive contamination in the workplace was co

to the process operations and was considered to be of significant concern only if it gave rise to high dose rates or contributed (by way of resuspension) to high airborne concentrations of radioactive material that could be inhaled. In June 1955, health physics identified that contamination levels in C-410 were higher than at any previous time. Excessive amounts of powder were present and the settling of UF<sub>4</sub> on the west mezzanine floor amounted to a green film that was noticeable even after the floor had been swept. Similar conditions and findings were noted in various health physics inspection reports and surveys through the 1960s and 1970s. The recurring nature of these findings from health physics inspections indicates that corrective actions were not taken to minimize these conditions or were ineffective.

### **Personal Protective Equipment (Information from Phase II Investigation)**

The use of PPE, and particularly respiratory protection equipment and coveralls, was inconsistent at PGDP. As early as 1952, the Health Physics and Industrial Hygiene Department recognized the potential hazards associated with personnel contamination and instituted measures to attempt to control potential exposures, including regular work area radiological surveys to determine the levels of personnel and clothing contamination. These surveys clearly indicated significant levels of radiological contaminants on hands, clothing, and shoes.

In several Plant areas, frisking devices were installed to allow personnel to self-monitor for radiological contaminants after hand washing before lunch and at the end of shift. Several thousand survey records for the period 1952 to 1956 indicate that significantly less than 1 percent of the personnel performing self-monitoring activities identified contamination on their hands. No routine survey program was established for clothing or shoes. On January 1, 1957, the Health Physics and Industrial Hygiene Department issued a letter to all division superintendents stating that workers did not need to wash their hands before eating to avoid concerns with radioactive contamination. Shortly after this letter, the use of hand counters was discontinued at the Plant until the 1980s.

The Health Physics and Industrial Hygiene Department issued memorandum to Paducah management entitled "Hand Contamination," evaluated entry pathways for U into the body. The conclusions presented in the memorandum were based upon studies at AEC and research facilities. For inhalation of U, the memo concluded, "smoking with contaminated hands is not a significant factor in U exposure." For ingestion, the memo stated, "Animal feeding experiments showed that insoluble compounds of U may be ingested in relatively large amounts without hazard." Similar conclusions were associated with injection of U into the skin of the hand. In 1958, Health Physics and Industrial Hygiene management recognized that major portions of the beta radiation exposure to workers resulted from contaminated coveralls. The Health Physics staff estimated exposures and added that to personnel exposure records. Typical annual additional skin doses due to contaminated coveralls were recorded in the 500 to 800 mrad range.

The Health Physics and Industrial Hygiene Department was actively involved with contamination control issues associated with the use of personal clothing in process areas. Following a 1956 review of the C-720 Electrical Shop, Health Physics and Industrial Hygiene stated that "Nothing was found which could be considered as detrimental to the health of the men working in this shop or to their families as a result of contamination being carried home on shoes or other clothing." In July 1957, management directed that personal clothing would be used on all work in the C-720 Control Valve Shop. However, evidence suggests that Paducah personnel routinely exceeded personal clothing contamination limits without corrective actions being taken by management. Health Physics surveys in the C-720 Control Valve Shop measured personal clothing contamination levels up to 2.5 mrad/hour and 1,250 dpm alpha. Similar measurements were

identified in October 1957, with the stipulation that the use of personal clothing was approved as \_\_\_\_\_, considering that an exposure of 600 mrad per week in a year's time would exceed the maximum allowable annual contamination and exposure to non-\_\_\_\_\_ with home laundering of the clothing.

In 1967, Health Physics and Industrial Hygiene management presented a position paper to all acknowledged applications where PPE should be utilized to maximize skin protection, the paper not clothing contamination levels of 4,000 cpm alpha were expected during the work to be performed. I clothing was to be utilized. The paper also highlighted supervision's responsibility to determine when contamination clothing should be issued and offered the Health Physics and I Hygiene Department's support in conducting surveys and providing supervisors with facts and advice. Supervisors and foremen were never issued company type clothing, even though in many cases those personnel were exposed to the same radiological h

### **(Information from Phase II Investigation)**

The PGDP's Health Physics and Industrial Hygiene Department considered personnel exposures -enriched U compounds to constitute a chemical rather than radiolog only were the constituents of U compounds within the enrichment cycle hazardous (e.g., fluoride and acid compounds), but also heavy metal poisoning could result from exposures to significant -enriched U. Consequently, re instituted to minimize personnel exposures to these contaminants. In general, the respiratory protection program utilized two basic types of respiratory protection equipment, the MSA It mask to minimize personnel exposures to dust-contaminants, respectively.

As early as 1953, Paducah management was aware that feed made from recycled reactor fuel processed through the enrichment cascade contained trace quantities of recognition of the potential for personnel exposures to these contaminants. However, at least initially the respiratory protection program and health physics surveys and monitoring did not nants. It was not until 1957 that the Health Physics and Industrial Hygiene Department discovered, during surveys, that Np had also entered the process stream from the reactor return feed materials.

During this period, the Health Physics and Industria collected air samples throughout the site. Sample records indicated that airborne contaminants, noted as alpha contaminants, exceeded the MAC. In many cases, after the fact, Health Physics e personnel routinely recommended the use of respiratory protection devices for specific tasks with identified high airborne radioactive material concentrations. recommendations, they were not always implemented.

In September 1953, urine bioassays for personnel involved in ash receiver handling operations respiratory protection equipment during these activities. As a result of this determination, the

site's Health Physics and Industrial Hygiene Department recommended suspending the practice of transferring ash receivers to drums as a means to reduce potential airborne Pu levels.

In 1957, radiochemical analysis of impurities from wet chemistry processes at the site revealed the presence of both Pu and Np. Further study concluded that the contaminant was confined to the chemical processing areas of the Plant. However, during Health Physics surveys in the Weld Shop in 1957, unusually high alpha contamination levels were detected on large diameter process piping. Records indicate that no visible U was present on the work piece, even though high smearable alpha contamination was detected. Radiochemical analysis of swipe samples indicated that 50 percent or more of the alpha activity on the work piece was due to <sup>237</sup>Np. This finding resulted in recognition that the entire cascade was contaminated with Np, and studies were conducted to determine which jobs presented the highest potential for exposure.

Many jobs were assessed for potential Np exposures. The Health Physics and Industrial Hygiene Department concluded that the disassembly of converters presented the highest exposure potential for Np exposure. Although the record indicates that dust respirators were used during converter work, elevated air sample results clearly indicated that airborne Np contamination presented a serious personnel exposure problem. Additional control measures were evaluated and implemented, including the use of ventilation systems and wetting of surfaces to reduce dust dispersion. When equipment size or configuration precluded the use of other control measures, records indicate that the use of air-supplied hoods was recommended.

The Health Physics and Industrial Hygiene Department actively promoted the use of respiratory protection devices in areas with high potential for airborne and/or chemical contaminants. The Health Physics and Industrial Hygiene Department routinely interacted with operations management and workers to advise on the use of respiratory protection equipment and provide counsel on the types of work that would normally require respiratory protection. However, the Health Physics and Industrial Hygiene Department did not have the authority to direct the use of respiratory protection. Consequently, respiratory protection was not always utilized when high levels of airborne contaminants were present. For example, a Health Physics and Industrial Hygiene Department quarterly report for the first quarter of 1959 reported that continuous air samples collected near the Np recovery operation in C-710 averaged slightly above the MAC assumed for Np. Later analysis indicated that 29 percent of the alpha activity was attributable to Np. There is no indication that respiratory protection was used during these activities. Urine samples collected and sent to ORNL for analysis tested positive for Np.

Work was routinely conducted without the benefit of respirators on open cascade components in process buildings, maintenance and refurbishment work, and waste handling activities, which were known to contain TRU compounds. Respiratory protection was not always used during UF<sub>6</sub> releases in process areas, and it was common for operators or Operations supervisors to enter the area of an active UF<sub>6</sub> release without respiratory protection or other PPE in order to stop the release.

The Health Physics and Industrial Hygiene report for January 1962 indicates that urinary excretion rates had steadily increased over the past several years, to the point that some personnel were excreting as much as 3 dpm/24 hour specimens. By current standards, the dose represented by this excretion rate would be well in excess of regulatory limits. This report also notes that the time-weighted average airborne Np alpha activity in the breathing zone of personnel disassembling converters from C-337 had increased and was 237 dpm/m<sup>3</sup>, with 90 percent of the alpha activity in the deposited dust on the equipment coming from Np. Controls included additional vacuuming and the use of air-supplied hoods instead of dust masks.

There is evidence that as late as 1973, inconsistencies in the use of respiratory protective equipment to employees allowed workers to choose whether to use a respirator, and what type, based on their perception of odor or visible fumes in the work area. It is only recommended that personnel leave the area of air contamination when necessary to obtain proper radiological hazard mentioned in the respiratory protection guidance, even though during the TRUs) were fed to the cascade.

## 2.6

### TRU or FP Constituents

During its first 40 years of operation, PGDP experienced numerous operational upsets, releases, events were very inconsistent and infrequent until the initiation of DOE's formal occurrence release of UF<sub>6</sub> process system during system upgrade work, equipment failures, and maintenance activities; equipment disassembly during shop maintenance activities in C-720.

Several evaluation reports of UF<sub>6</sub> releases and their effects, as well as other site documents, of UF<sub>6</sub> releases, each in excess of 10 pounds of U. However, reviews of released references to many hundreds of releases of varying sizes (described often only as minor, large, or required medical examinations and bioassay. Many health physics reports indicated that these quarterly progress reports to the AEC, which was the regulatory agency at that time (Phase II Investigation).

There were at least a minimum of 100 pounds of U. A 1960 event released approximately 6,800 pounds and a 1962 event releasing approximately 3,400 pounds. As better equipment was installed and upgrade work ended, operational practices improved and the number and quantity of UF<sub>6</sub> decreased significantly. In the 1980s, reported releases were on the order of one to five per month and were measured in grams instead of pounds. The bioassay decreased from 30 or more per month in the 1950s to one to six per year in the 1980s

-310 in 1956, overexposure of two maintenance mechanics to beta radiation, and an explosion and fire in C- releases affecting groundwater included a spill of 17,000 gallons of diesel oil migrating as far as 2 miles from the site boundary via surface water and the identified TCE leakage from C- reported as a result of Plant events: an explosion and fire in C- electrical maintenance trainee in 1977, and the suffocation of an operator in the collapse of a coal a worker who did not return to work.

## **2.7 Environmental Monitoring and Records**

### **Environmental Management Practices**

PGDP operations have resulted in the release of a variety of contaminants into the environment through stack and diffuse air emissions; discharges through sewers into lagoons, local ditches, and streams; accidental releases; and past waste disposal practices such as the burial of low-level and hazardous waste.

The primary mission of the Plant involved the enrichment of U to support defense and commercial nuclear industries. The U used in the Plant was obtained both from commercial industries and from the recycle of reactor tails through separating irradiated fuel and targets. These reactor tails contained trace levels of TRU and FP, which were introduced into the enrichment system and the resulting waste materials. U was the largest contributor to environmental contamination. Because U was a valued commodity, U releases and transfers were minimized from the start of Plant operations in 1952. A variety of chemicals were used directly in the feed production and enrichment processes, or used to in support operations such as cooling water treatment and cleaning.

Requirements relating to the release of chemical and radionuclides into the environment were limited in the early years of Plant operations. The AEC established allowable limits for the release of radionuclides into the environment, but Federal and state agencies had few restrictions on discharge and disposal activities until the late 1960s. Releases from U.S. industrial operations during the 1950s and 1960s, including those at Paducah, were significant. Past PGDP operations resulted in a significant environmental degradation in the vicinity of the Plant due to the accumulation and transport of contaminants associated with past disposal and spill sites as well as release and migration of contaminants to local streams and groundwater. DOE submitted a RCRA Part B permit on February 8, 1985; this permit and a RCRA Hazardous and Solid Waste Amendments permit were effective on August 19, 1991. In May 1994, PGDP was listed on the National Priorities under CERCLA, and in February 1998, the DOE, EPA, and the Commonwealth of Kentucky entered into a Federal Facility Agreement for environmental remediation. On February 20, 1992, DOE and EPA entered into the Uranium Enrichment Federal Facility Compliance Agreement that regulated PCB removal and disposal at PGDP.

### **Waste Management**

Construction and operations at PGDP generated a wide variety of waste and scrap materials beginning in the early 1950s. An integrated waste management program did not begin at the Plant until the early 1980s. Before the establishment of this integrated program, each organization at the Plant disposed of its own waste. The Maintenance Department provided support by operating a number of common disposal sites.

The formation of an integrated program began in response to a December 1978 report by the site Environmental Control Department on disposal of solid waste (including radioactive and hazardous waste). This report stated that the Plant was not meeting current and planned solid waste regulations. In addition to the recommendations for better management of existing facilities and the need for additional facilities, the report recommended that specific individuals be made responsible for operation, maintenance, record-keeping, and planning of solid waste storage and disposal areas. The resulting organization, the Material Terminal Management (MTM)

Department, implemented the integrated waste management program by gaining control of the waste management facilities and developing waste management procedures for the Plant.

### **Solid Waste Disposal**

During construction of the original Plant, the prime contractor established an inert disposal site for construction rubble north of the Plant. Over time, this site continued to be used for disposing of construction materials. As the Plant became operational and generated hazardous and radioactive waste materials, contaminated materials were introduced into this disposal site, including contaminated roofing material and concrete, asbestos, and chemically treated wood from the cooling towers. On the southwest side of the Plant, a borrow pit was used to dispose of ash from the Plant's coal-fired steam plant, which was subsequently designated as the C-746-K landfill.

Over time, these two sites apparently evolved into landfills not requiring permits according to Commonwealth of Kentucky regulations; the Maintenance Department operated the landfills. The limit established during early site operations for radioactive material in these areas was 2 pounds of U per ton. For depleted U, the limit would correspond to a volumetric concentration of approximately 333 pCi/g or 670 pCi/g for natural U. Floor sweepings were disposed of at these landfills. Since process materials, including green salt and yellowcake, were routinely present in large quantities on floors and equipment in some buildings, it is possible that these radioactive materials, in much higher concentrations than allowed, were inappropriately sent to these landfills.

Within the Plant's security fence in the northwest corner, a 30-foot-high ramp and pit arrangement, known as the teepee, was used to burn combustible waste. As an aid to combustion, waste oils were added; however, these oils were not controlled and they were likely contaminated with solvents and PCBs. This operation continued until December 1, 1967, when air control regulations for open burning at disposal sites required termination. At that time, these waste streams were sent to the coal ash disposal site.

Although landfills were on government property and patrolled by Plant security, the public could access these areas. Some members of the public routinely retrieved scrap wood and others used construction items from the inert disposal site, starting during Plant construction and continuing into the 1970s. At the 746-K landfill, for example, redwood with brass bolts from the cooling towers and used wood paneling from Plant offices attracted salvaging from the public and possibly workers (Phase II Investigation). Limited controls had been established on disposal of material from the cascade and other process and operations buildings in order to keep highly contaminated items from going to the landfill. However, when surveillances were conducted at the landfill in later years, such items would occasionally be identified, indicating weaknesses in the implementation of management controls.

Maintenance workers operated these disposal sites during the day, implementing verbal guidance from their supervisors. These workers used bulldozers and other heavy equipment to compact and dress the working areas. Since their equipment did not have closed cabs, the workers may have been exposed to both unconfined asbestos and ash from the coal-fired steam plant. As the Plant's heavy equipment operators, these workers also hauled construction rubble to both the landfills and the inert disposal areas around the Plant, including parts of what is now the Kentucky Wildlife Area. Concrete rubble and debris, some with radioactive contamination, was sent offsite to areas in the former KOW. The limited space available for disposal within the Plant security fence probably affected the decision to discard these materials on the KOW.



In the early 1980s, additional controls were implemented at the landfills. These controls eventually included controlled access to the landfills, waste acceptance criteria, record keeping, and licensing both the landfill and the operators with the Commonwealth of Kentucky. Controls were also applied to waste generators. Segregated dumpsters for both non-hazardous and radioactive wastes were acquired, and procedures and guidance on acceptable disposal practices in the Plant's sanitary landfills were established.

### **Radioactive Waste Management**

Radioactive waste management has been evolving since the 1950s. In April 1953, efforts were initiated to reduce the spread of contamination by using drums designed for disposal in work locations known for generating highly contaminated waste. Operating logs in C-340 from 1958 discuss using a supply of scrap drums from the holding pond for packaging black oxide rather than putting the oxide in dumpsters (Phase II Investigation). Actions to segregate these wastes from the Plant's other waste streams resulted in establishing radioactive disposal sites. Although several small sites were used for special disposal activities, including contaminated aluminum and a modine trap, the Plant had three main radioactive disposal sites:

- C-749 U Burial Ground. Used from 1957 to 1977, this site primarily contained pyrophoric U metal in the form of sawdust, shavings, and turnings covered in oil. The total amount of U placed in this site is approximately 540,000 pounds (Phase II Investigation).
- C-340 Drum and Contaminated Burial Area. Used from the late 1950s until the mid-1970s, this area received C-340 U powder scrap. In the 1950s, 50 to 75 drums were emptied into a pit 10 feet by 20 feet, and 7 feet deep. In the 1970s, two more 7-foot-deep pits were used for disposal of contaminated metals and equipment (Phase II Investigation).
- C-404 Solid Radioactive Waste Disposal Area. This was the primary disposal site for radioactive waste at the Plant. This area was constructed as a holding pond for C-400 liquid waste, but in early 1957, it was converted to a solid waste disposal area. The pond was 380 by 140 feet, with 6-foot-high dikes. By 1977, approximately 6,400,000 pounds of U had been drummed and placed in the holding area. Waste streams included incinerator ash, contaminated alumina, highly contaminated roofing waste, and gold recovery sludge. This area continued in use into the mid-1980s. Subsequently, this area was determined to contain sludge that was also hazardous, thus requiring closure under RCRA in 1987 (Phase II Investigation).

After the formation of the MTM Department, radioactive waste disposal on site rapidly decreased. In 1978 and 1979, the amount of disposal was 330,690 pounds per year; in the 1980s, the average was 18,000 pounds per year (Phase II Investigation). As a result of not burying radioactive waste on site and restrictions for offsite disposal, the site experienced a large buildup of contaminated waste and scrap.

### **Management and Disposal of Scrap and Surplus Materials**

Large volumes of scrap metal and surplus materials were generated during construction, maintenance, and facility upgrade activities at PGDP. These materials were either managed as waste for disposal or stored and managed as a commodity for resale. Much of the material was contaminated, and large volumes have been disposed of on site. Additionally, large volumes of

scrap metals remain in outside storage pending resolution of policy issues associated with the resale of contaminated materials.

As part of scrap metal activities at the Plant, generators were responsible for requesting disposal containers for contaminated and clean scrap metal and then inspecting these containers to determine proper usage. The Material Services Department established and maintained approved scrap yards, and the Maintenance Services Department collected and transported the containers. In addition, special responsibilities for classified scrap were defined in a standard practice procedure. This procedure, dated July 24, 1969, identified four scrap yards: C-746-F for buried classified material; C-746-E for contaminated material; an unclassified burial yard; and C-746-C for clean material (defined as less than 1000 alpha c/m/100 cm<sup>2</sup> and less than 0.3 mrad/hr beta-gamma). This 1969 procedure also required supervisors to determine whether contamination was sufficient to warrant recovery at the C-400 decontamination and recovery area. If not, the scrap was sent to one of the four disposal yards. Source areas for scrap metal included the C-340 metals plant, the cascades, C-410 and C-420 feed plant, the C-720 fabrication and maintenance shops, and the laboratories. Part of the scrap was buried directly (classified material). As the disposal areas were filled, non-classified material began to accumulate in aboveground piles that still exist. These yards and disposal locations are now identified as SWMUs for investigation and possible cleanup under the current restoration program.

Some of the metal components, equipment, and vehicles at PGDP had significant inherent value, including a large amount of material that was considered scrap as a result of upgrades or replacement of equipment and process piping. Consequently, management wished to sell to interested parties as much of this material as was possible. Based on Plant health physics records, it was clearly understood as far back as the 1950s that "contaminated" material above certain limits could not be sold or released to the public (Phase II Investigation). Therefore, the handling and disposal of scrap materials was subject to the corporate procedures described above. While contamination limits and specific categories changed over the years, scrap was required to generally be categorized into one of four groups: classified scrap, unclassified clean scrap, unclassified contaminated scrap, and unclassified non-metal trash. Dumpster pans were provided for each of these categories wherever significant quantities of scrap were generated. Line supervisors were responsible for ensuring that employees segregated all scrap materials appropriately. Once full, these dumpsters were hauled to a designated location. The material categorized as clean unclassified scrap was taken to the C-746-C clean scrap yards for placement and preparation for public sale.

Health Physics surveys of the clean scrap materials were primarily cursory, consisting largely of periodic inspections and spot-checking of suspect materials in the clean scrap yards based on process knowledge. Vehicle floorboards and seats were also spot-checked before sale to the public, but the process was informal and was not required by procedure. The Health Physics and Industrial Hygiene Department was on the distribution for notices of public sales and was aware of their responsibility to survey "suspect" items to be sold. Documentation that proper radiological surveys were performed was not consistently maintained until the late 1980s, when the Health Physics and Industrial Hygiene Department began to place more emphasis on maintaining formal records for radiological release of material and equipment from the site.

It is possible that contaminated items were released to various parties during public sales. The site was doing a less than adequate job of segregating clean from contaminated scrap and that contaminated scrap was often found in clean scrap locations. In May 1976, a health physics inspection of the C-746C "clean" scrap yards identified a number of prohibited contaminated items, most notably a 30-gallon drum of U metal shavings (Phase II Investigation). A Scrap

Handling Committee was formed in mid-1976 to study PGDP solid waste disposal problems, including the issue of segregating contaminated scrap.

### **Liquid Effluents**

Liquid effluents were historically released in a number of ways, including via the sanitary sewage and storm water drainage systems. Eventually, effluent material that was not otherwise held up or recovered through wastewater treatment and recovery systems flowed to one or more of the various site outfalls and ditches and then into either the Big or Little Bayou Creeks, which ultimately discharged to the Ohio River.

In the early 1970s, the Clean Water Act established the NPDES, which administered effluent limitations and water quality requirements for chemical releases. These programs could be administered by the states after Federal authorization. In Kentucky, these were known as KPDES permits. KPDES permits only requires reporting of radiation levels, no limits. The first one was issued for the recirculating cooling tower blowdown water. Subsequently, a total of 18 outfalls were permitted at the site. Liquid effluent discharge limits for radionuclides were not specifically promulgated by EPA, but were always required and published under the AEC and ERDA regulations and later DOE orders as MPC or RCGs in water. Despite the discharge restrictions, enough radionuclides have been released to create legacy environmental contamination. The existence of legacy contamination has been confirmed through environmental sampling data.

The most significant liquid radiological effluent source was the C-400 decontamination building. This building contained a variety of systems and processes for isotopic recovery and decontamination of process equipment and scrap metal, as well as the sitewide laundry. Given the nature of operations in this facility, managing the various types and quantities of liquid wastes generated was a significant challenge. These wastes included TCE from degreasing operations, contaminated liquids from cleaning operations, and various contaminated raffinate solutions from U, Np, and <sup>99</sup>Tc recovery operations. For radionuclides, essentially all isotopes at the site were present in various portions of this facility and in its liquid waste streams, including U, Np, Pu, Th, and <sup>99</sup>Tc.

U recovery operations in this building were used to recover valuable U materials and also to reduce the U concentration in cleaning liquids to acceptable levels before release. Np and <sup>99</sup>Tc recovery campaigns were also conducted at various times during Plant operations. Liquid effluents from these operations and others that generated contaminated liquids were sampled before being released to drainage systems. If the applicable limits were not met, the material was either put in drums and stored or routed back through the U recovery process. Liquids that met the discharge limits were released to the North-South Diversion Ditch and outfalls, depending on the piping sequence. In 1972, Union Carbide reported that from 1956 to 1970, the U recovery system discharged a total of 4000 grams of Np and 191 grams of Pu to the environment (Phase II Investigation).

In 1963, the AEC authorized a request by the site to release Th-, Np-, and U-contaminated raffinate solution being stored in drums to the Ohio River via a diversion ditch (Phase II Investigation). The request stated that the discharge would be controlled to keep the concentration of the materials in the river below permissible limits. The request was granted by the AEC, effectively allowing the point of compliance for liquid effluents to be the Ohio River, rather than local ditches and streams. This decision may have been a misapplication of AEC regulations concerning maximum permissible concentrations of liquid effluents in unrestricted areas. This

type of approach has contributed to elevated isotopic concentrations of U, Th, TRUs, and FP found in ditches and outfalls both on and near the site today.

The outdoor storage and placement of contaminated waste and scrap that began in the late 1950s (e.g., Drum Mountain and scrap yards) has continuously contributed to the spread of contamination through surface water runoff. Contaminants settled in onsite ditches and streams. As a result, in the late 1980s efforts were undertaken to characterize and plan for remedial measures to address these contaminants. Limited removal and access controls were established in the 1990s.

From the beginning of PGDP operations, the C-615 sewage treatment plant treated sanitary wastewater (sewage and sink wastes) from process and support buildings. Radiological components of treated water caused the sewage sludge to be contaminated with U. Subsequently; this material was unknowingly spread at various locations at the site, creating contamination control problems. In 1977, the C-616 wastewater treatment plant came on line. Major liquid effluent streams that fed into the North-South Diversion Ditch, were then routed by a lift station to the 616 facility, resulting in a significant improvement in water quality in local streams.

### **Atmospheric Releases of Radioactivity and Fluorine/ Fluorides**

Radioactive and fluorine/fluoride air emissions to the atmosphere began with startup in 1952 and have continued to the present from USEC operations regulated by NRC. The air emissions from the site were from process stacks, diffuse and fugitive emission sources, accidental releases, and a limited number of planned releases.

### **Stack Emissions**

The site did not perform stack monitoring until the mid-1970s, so the actual quantities of radionuclides released to the environment from routine operations before that time are unknown. From 1959 to 1974, the air emission reports consisted of ambient air monitoring. Starting in mid-1960, continuous ambient air samples were taken at four locations at the perimeter fence and were analyzed for alpha and beta activity to provide input for estimation of annual ambient air concentrations. In 1961, four additional ambient continuous air samplers were installed one mile outside the perimeter fence. Since stack emissions were not measured from 1952 to 1974, the Health Physics and Industrial Hygiene Department estimated emissions based on Plant operations. Interviews indicated that the estimates were probably within a factor of two but could be off by as much as a factor of five (Phase II Investigation). It was not clear whether accidents that occurred during this period were considered in the emission estimates.

The first environmental report indicating stack emissions of U and <sup>99</sup>Tc were prepared in 1976 for the 1975 calendar year. For the years after 1975, the Environmental Reports also reported annual discharges to the atmosphere based on stack measurements.

The site, using available information, estimated that approximately 60,000 kg of U was released to the atmosphere between 1952 and 1990. Of the total, approximately 75 percent was estimated to have been released before 1965 (Phase II Investigation). Most of the estimated releases were attributed to the C-410 feed plant and the C-340 metals plant. C-410 was shut down in 1962, reactivated in 1968, and finally shut down in 1977; C-340 was first shut down in 1964, reactivated in 1968, and finally shut down in October 1973. When these plants were shut down, estimated air emissions from the site were greatly decreased.

### **Accidental Releases**

A number of accidental releases to the atmosphere have occurred at PGDP. There have been 15 accidents that released more than 50 pounds of  $\text{UF}_6$  from cylinders. Approximately 300 material releases (most of them accidental) occurred from July 1, 1952, to July 1, 1972 (Phase II Investigation). These included releases to the atmosphere and some discharges to water. Sixty-nine (excluding routine stack emissions) were probable airborne releases of more than 10 pounds of U each (Phase II Investigation).

From 1960 to 1974, heavy reliance was placed on ambient air samples for assessing impact on the public. However, ambient air samples only measure plumes at ground level. Lofted plumes might not be measured, depending on the meteorological conditions. Plume lofting is expected during accidental releases of  $\text{UF}_6$ , since the reaction between the  $\text{UF}_6$  and water vapor releases heat. The expected plume lofting was observed during two accidental releases on May 20, 1958. An attempt was made to sample the plumes downwind from the Plant. The first plume was observed to intersect the ground, while the second plume remained elevated (Phase II Investigation).

### **Diffuse and Fugitive Emissions**

Diffuse and fugitive emissions were generally not calculated for the site from 1952 through 1990. A limited set of data exists for releases during the mid-1950s from some processes, such as U metal pickling, smoking ash receivers, and drum dryer exhaust (Phase II Investigation). Workplace air samplers and contamination on roofs and ground in the site area point to the occurrence of unmonitored releases. One example is the C-404 Holding Pond. U-contaminated water was originally piped to the pond, and in 1957 the pond was turned into a solid waste burial area. A ramp was later constructed to reduce dust emissions from the area. There was no evidence that diffuse and fugitive emissions were substantively included in release inventories and subsequent public dose calculations. Also, even though diffuse emissions of TRUs would have occurred during pulverizing of the feed plant receiver ash, no estimates of these emissions were found.

### **Planned Releases**

Four planned atmospheric releases of  $\text{UF}_6$  occurred at PGDP: two 4.4 kg releases in 1955 and two 0.68 kg releases in 1974 (Phase II Investigation). These releases were designed to model plume behavior from a surface release and were followed by an additional series of tests where approximately 160 grams of  $\text{UF}_6$  was released at ground level directly into the atmosphere (Phase II Investigation). Finally, six releases occurred in the 1975-1976 timeframe, involving a total of approximately 1 kg of  $\text{UF}_6$  (Phase II Investigation).

## **3.0 Recycled Uranium Mass Flow**

Materials received at PGDP processing as plant feed could come in many forms, i.e.:  $\text{UO}_2$ ,  $\text{UO}_3$ ,  $\text{U}_3\text{O}_8$ , and  $\text{UF}_4$ . The feed conversion process could utilize any of these forms, although  $\text{U}_3\text{O}_8$  would require more complex pre-processing. All material received as oxides from Savannah River and Hanford were immediately identified as RU. In determining the mass flow of recycled material, some basic assumptions had to be made on identifying some portions of feed since not all materials were consistently identified as recycled U. Incoming materials with assays less than 0.3%  $^{235}\text{U}$  were eliminated from consideration as feed material. Masses of the material less than 0.1 MTU contribute little to the overall equation for TRUs. These materials were entered into the database (see Appendix B) in order to achieve a reasonable mass total, but calculations on total

amounts were made on materials rounded to the nearest MTU. Materials such as  $\text{UO}_2$  with total amounts received of less than 5 MTU were not considered in the mass flow because of negligible contributions to the total. A substantial amount of total material received by PGDP was of normal material and not from recycled sources. Because of accounting processes, some of this material was described as “depleted” or “enriched”, when its deviation from “normal” material was minute ( $0.001\% \text{ }^{235}\text{U}$ ). Only feed material listed as normal with assay less than  $0.68\% \text{ }^{235}\text{U}$  and greater than  $0.72\% \text{ }^{235}\text{U}$  were judged to be of recycled origin.

Since analytical data on impurities present in feed materials are extremely limited or inaccessible, especially in initial operation years, correlation of analytical data with specific shipments and fiscal year summaries could not be achieved. There were however available data in KY-L-411 on 128 analyses of Savannah River and 238 analyses of Hanford materials spanning most of the years of interest. Although no direct linkage could be made on lots, these data points allow a FY average to be calculated. It is felt that this will provide a better picture of year-to-year receipts. It was still necessary to abstract some data from the Smith report to fill data gaps.

Movements of the impurities from feed to final product have also been described in previous studies. The results of these studies were used extensively in deciding where and in what amounts these impurities would transfer or reside (KY-L-936,  $^{99}\text{Tc}$  and TRU Summary). From the feed preparation, 1%, of the Pu, 75% of the Np, and 95% of the  $^{99}\text{Tc}$  present in oxides would pass as feed into the  $\text{UF}_6$  cylinder. The ash and vacuum dusts would contain a substantial amount of Np (25%) and Pu (>95%). Pu transferred into the cylinder, because of its reactivity, would tend to stay in the “heel” with only a limited amount ever introduced into the cascade (<0.1%). Approximately 25 % of the Np in the cylinder would be fed to the cascade.  $^{99}\text{Tc}$ , on the other hand, would transfer with the  $\text{UF}_6$  and be fed, (85%), ultimately to the cascade. (All percentages listed are percentages of the amount in the original feed material (oxides, chiefly).  $^{99}\text{Tc}$ , because of its mass, would migrate in a short time to the top of the cascade and reside there in a “bubble”. It would then be withdrawn in small quantities along with product enriched  $\text{UF}_6$ .

Previous studies presented arguments for the validity of their assumptions. In using the assumptions, decisions were made based on the latest available information and choices made on best applicability. Overall validity of summation data is felt to be an accurate total depiction of flow. Mass flow of recycled material required examination of feed materials by type and source.

Reactor tails (RU) materials in the form of  $\text{UO}_3$  were received at the PGDP from Hanford, FMPC (NLO), or Savannah River facilities from fiscal year 1953 through fiscal year 1989 (excluding fiscal years 1965 through 1968, 1976A, and 1977 through 1985). A small amount of oxides were received from Harshaw and ORGDP and are listed as miscellaneous  $\text{UO}_3$ . A fiscal year summary of shipments and receipts of the oxides by source is presented in Table 3.0-1.

**Table 3.0-1, Recycled  $\text{UO}_3$ , MTU**

FY	$\text{UO}_3$ fr. S.R.	$\text{UO}_3$ to S.R.	$\text{UO}_3$ fr. Han.	$\text{UO}_3$ to Han.	$\text{UO}_3$ fr. Misc.	$\text{UO}_3$ to ORGDP	$\text{UO}_3$ fr. FMPC	$\text{UO}_3$ to FMPC	Net RU R'c'd
53					68				68
54			2170		152	1.8			2320
55	283		2647			3.8			2926
56	171		3972		0	21.0			4122
57	565	0.5	5428	2.4		15.8			5974
58	229	0.1	6104	2		8.6			6322
59			5112	2.1	21	10.0			5121

FY	UO <sub>3</sub> fr. S.R.	UO <sub>3</sub> to S.R.	UO <sub>3</sub> fr. Han.	UO <sub>3</sub> to Han.	UO <sub>3</sub> fr. Misc.	UO <sub>3</sub> to ORGDP	UO <sub>3</sub> fr. FMPC	UO <sub>3</sub> to FMPC	Net RU R'c'd
60			5151	1.8					5149
61	81		6001	2.9	10				6089
62	1546	1.8	5630	4.1	0	15.2			7159
63	1344	1.2	5775	4		35.5	0.3		7079
64	1603	1.1	4178	2.3		7.4			5770
65		0.3							0
66									
67			14432						14432
68									
69	2812		3537				0.2		6349
70	597				14				611
71	34		714				761	0.1	1509
72	-34		3072				1779	0.5	4817
73			556				386		942
74								0.2	-0.2
75									
76								0.5	-0.5
77							1002		1002
78									
79								85.6	-86
80									
81									
82								665	-665
83								397	-397
<b>sub t.</b>	9231	5	74479	22	265	119	3929	1149	
<b>net</b>	9226		74457		146		2780		
<b>Net RU UO3</b>									<b>86609</b>

\*Shipments indicated above of less than 3 metric tons U were primarily returns of material in shipping containers. Misc. category is defined as receipts from Harshaw and ORGDP. Harshaw receipts are only in FY-53 and FY-54. In addition, 10 MTUs from Hanford in 1967 were of a 0.2 assay and as such were not material for feed but are included in the total of 14,432 for material balance reconciliation. This material was subsequently shipped to FMPC in 1983. Included in the FMPC totals is material of approximately 0.72 assay and assumed to be RU material since it was coded as RU material fed to the cascade.

In addition, significant quantities of recycled U were received at Paducah in the first few years of plant operation as UF<sub>6</sub> from the feed plant at ORGDP. Minor amounts were received from Portsmouth. Other small quantities of recycled U were received from foreign sources. Some of this material was never fed to the cascade and remains in storage (Commurhex). Receipts and shipments of RU UF<sub>6</sub> are summarized in Table 3.0-2

**Table 3.0-2, Recycled Uranium as MTU UF<sub>6</sub> (NMC&A codes 24 & 30)**

<b>FY</b>	<b>UF<sub>6</sub> From ORGDP</b>	<b>UF<sub>6</sub> To ORGDP</b>	<b>UF<sub>6</sub> From Ports</b>	<b>UF<sub>6</sub> To Ports</b>	<b>UK UF<sub>6</sub></b>	<b>Commurhex</b>
53	1091	1.1				
54	1644					
55	687	4.3	0.445	121.2		
56	2509	7	0.885	39.5	411	
57	2853	17		6.2	220	
58	881	12.7		57.8	372	
59	493	6.4		0.1		
60	574	1.9		0.1		
61	68	0.1				
62						
63						
64	0.6					
65						
66						
67						
68						
69		234		568		
70		335.6				
71						
72						
73						
74					74	
75						
76						
77						
78						
79						
80						
81						
82						
83						
84						
85						
86	485.7					75.7
87						165.6
88						223.4
<b>TOTAL</b>	11286.3	620.1	1.33	792.9	1077	464.7
<b>NET</b>		10666.2		-791.6	1077	464.7

Additional quantities of U as UF<sub>4</sub> and of possible reactor origin were received from FMPC, Y-12, and ORGDP. Receipts and shipments of this material are summarized in Table 3.0-3.



**Table 3.0-3, Receipts and Shipments of RU as UF<sub>4</sub>, MTU**

<b>FY</b>	<b>UF<sub>4</sub> from ORGDP</b>	<b>UF<sub>4</sub> to ORGDP</b>	<b>UF<sub>4</sub> from FMPC</b>	<b>UF<sub>4</sub> to FMPC</b>	<b>UF<sub>4</sub> from Y-12</b>
53	0.9				
54					
55	213.3	24.7			
56	117		9.3		16.1
57		1			10.2
58		140.4	5.5		
59		62.5			
60					
61	9.2				
62	2.3	1		68.6	
63					
64					
65					
66					
67					
68			30.8		
69			61.3		
70			7.8		
71			9.1		
72			2.2		
73					
74					
75					
76				109.1	
<b>subtotal</b>	342.7	229.6	126	177.7	26.3
<b>total</b>		113.1		-51.7	26.3

\*Net contributions of UF<sub>4</sub> are thus 87.7 MTU.

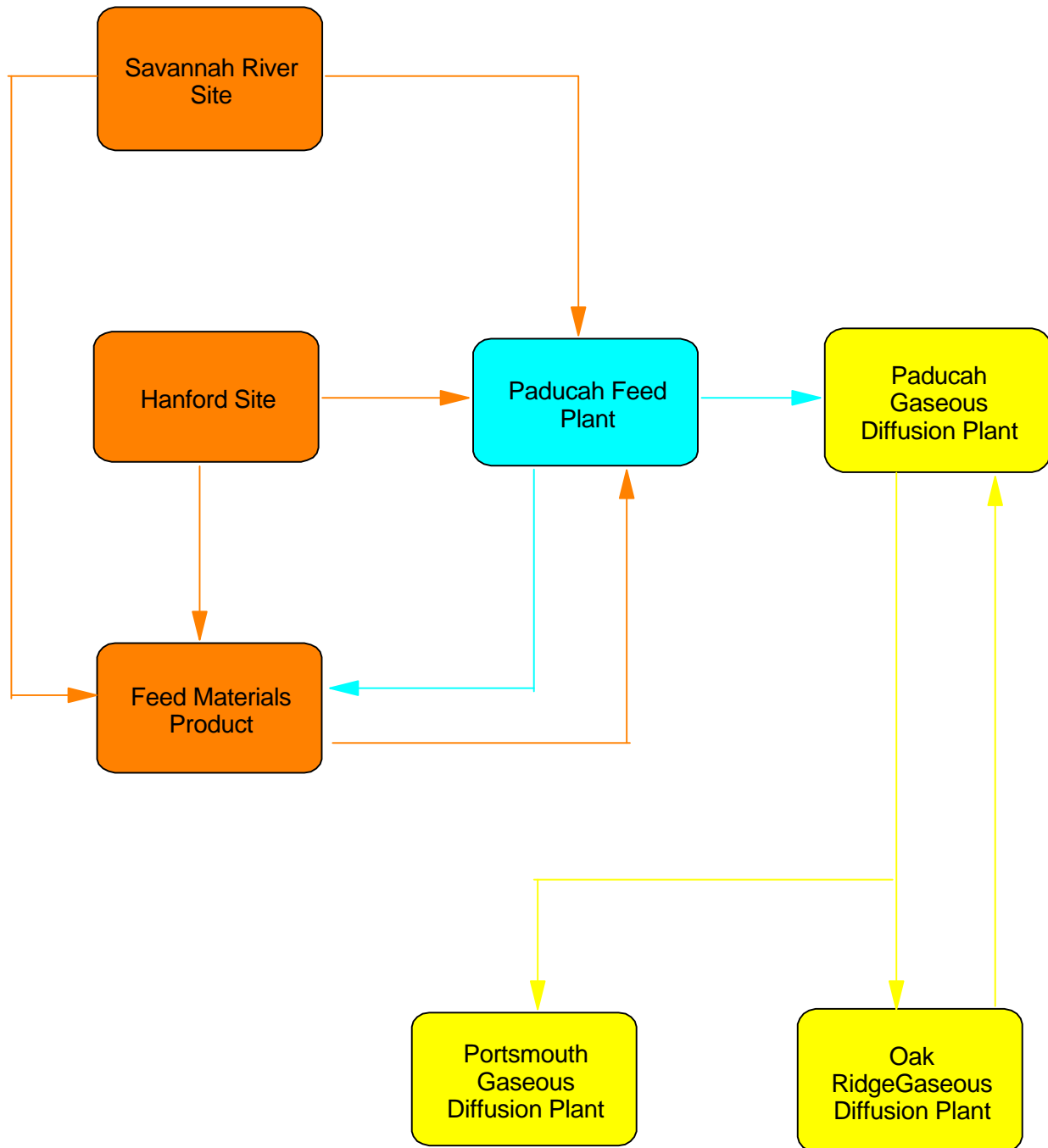
Contributions of U<sub>3</sub>O<sub>8</sub> to total net mass were also very small: 5 MTU from Dow Chemical, 1 MTU each from PORTS and ORGDP and 67 MTU from Y-12. Only a total of 138 MTU of U<sub>3</sub>O<sub>8</sub> were processed through the feed plant. Total contribution to the amount of impurities would have been very low.

### **3.1 Uranium Recycle Description**

Early in the operation of the first gaseous diffusion plant, uraniumhexafluoride feed was received in small batches (400 lb cylinders) from an off-site supplier. This made operation of the cascade difficult at high feed rates. Subsequently, a continuous feed processing facility was designed and constructed. This feed production facility had the capability to reduce UO<sub>3</sub> to UO<sub>2</sub> in the presence of hydrogen at elevated temperature, hydro-fluorinate the oxide with anhydrous HF vapor to produce UF<sub>4</sub>, and convert the UF<sub>4</sub> in a flame tower reactor in the presence of fluorine gas to produce UF<sub>6</sub>. The UF<sub>6</sub> gas was condensed and collected in large cylinders for feeding into the cascade. These early fluorination reactors produced “ash” from the incomplete reaction of the feed material and from small amounts of impurities that were originally present in the oxide that were non-reactive (such as U daughter products). A metal canister and filter were attached to each fluorination reactor to collect the ash in the canister and filter the UF<sub>6</sub> gas to remove

particulates. The metal canisters were removed periodically and cleaned to recover the unreacted or partially reacted U compounds. If the reaction were inefficient (containing a large amount of  $UF_4$ ), the ash material would be pulverized and reintroduced to the fluorination process. When the reaction was more complete, the ash was not sent through the fluorination process. Instead the ash was dissolved and processed through the wet recovery system, along with decontamination solutions and cylinder washings where the U was extracted as the nitrate, calcined and re-fed through the fluorination system. The ash recovery operated in this mode of wet recovery from 1953 until 1970. TRU impurities followed the raffinate in the extraction system. This enabled the loss to the environment noted in every reference to TRU materials handling at PGDP. When this reprocessing was discontinued in 1970, the ash was stored and eventually shipped to FMPC for U recovery. Two major shipments of ash were made to FMPC for U recovery. These shipments occurred in 1975 and 1980. The flow of materials between the various DOE sites is depicted in Figure 3.1-1, U Recycle.

**Figure 3.1-1, Uranium Recycle**



The feed preparation facility was ideal for receiving and processing of recycled U whose product form was  $\text{UO}_3$ . Approximately 84,000 MTU was received by PGDP from Hanford and Savannah River and miscellaneous sources as  $\text{UO}_3$ . An additional 44,000 MTU were received from FMPC, however, this material appears to be primarily natural U feed. [Approximately 980 MTU are assumed to be RU since the assay range of the U is other than natural. A additional 2948 MTU received are assumed to also be recycled material since the assay range is greater than normal (approximately 0.72%)]. Based on the current review of plant accountability records, recycled material was processed almost immediately following startup of the PGDP in FY-53 and continued through FY-64. A second RU campaign occurred from FY-69 until FY-74. Some recycled oxide was fluorinated at Oak Ridge for use as feed in Paducah. The percentage of the total feed to the PGDP represented by the RU varied greatly over the years with no RU fed in many years. At PGDP the amount was about 65 percent of total feed in FY-73. PGDP continues to store 1,267 MTU of RU (as  $\text{UF}_6$ ) on site. The PGDP uranium hexafluoride feed plant operated from 1953 through 1964 and 1968 through 1977 until it was shutdown (not operated for a four year period from 1964- 1968). RU was fed to PGDP cascade from 1953 to 1964, from 1968 to 1970, and from 1972 to 1976. An additional 7.3 MTU of Commurhex were fed from 1986 to 1989 as a result of cylinder transfer operations.

### 3.2 Uranium Receipts

Total U receipts will include U as  $\text{UF}_6$ ,  $\text{UF}_4$ , and  $\text{UO}_3$  as recycle uranium from all sources. This total represents the full amount of recycle uranium potentially available for feed to the cascade. Some of this material was not fed, not converted to feed material, stored on site, or shipped out to other sites. Total receipts are summarized in Table 3.2-1

**Table 3.2-1, Recycled Uranium Flow, MTU**

Materials Received		Materials Shipped				
FY	MTU $\text{UO}_3$	MTU $\text{UF}_6$	MTU $\text{UF}_4^*$	MTU $\text{UO}_3$	MTU $\text{UF}_6$	MTU $\text{UF}_4^*$
53	68	1091	0.9			
54	2322	1644		1.8		
55	2930	687.4	213.3	3.8	1.1	24.7
56	4143.2	2921	142.4	21	125.5	
57	5993	373	10.2	18.7	46.5	1
58	6333	1253	5.5	10.6	23.2	140.4
59	5133.1	493		12.1	70.5	62.5
60	5151	4		1.8	6.5	
61	6092.2	68	9.2	2.9	2	
62	7176.3		2.3	21.1		69.6
63	7119.3			40.7		
64	5781	0.6		10.8		
65				0.3		
66						
67	14432					
68			30.8			

<b>Materials Received</b>		<b>Materials Shipped</b>				
<b>FY</b>	<b>MTU UO<sub>3</sub></b>	<b>MTU UF<sub>6</sub></b>	<b>MTU UF<sub>4</sub>*</b>	<b>MTU UO<sub>3</sub></b>	<b>MTU UF<sub>6</sub></b>	<b>MTU UF<sub>4</sub>*</b>
69	6347.2		61.3		802	
70	12.2		7.8		335.6	
71	2072		9.1	0.1		
72	4852		2.2	0.5		
73	942					
74		74		0.2		
75						
76						109.1
77	1002					
78						
79				86		
80						
81						
82				665		
83				397		
84						
85						
86		561.4				
87		165.6				
88		223.4				
<b>Total</b>	87891.5	12829.4	495	1420	1412.9	407.3
<b>NET UO<sub>3</sub></b>						86471
<b>NET UF<sub>6</sub></b>						11416.5
<b>NET UF<sub>4</sub>*</b>						87.7

\* UF<sub>4</sub> with assays (% <sup>235</sup>U) less than 0.68 and greater than 0.72 assumed to be from reactor returns

### 3.3 Uranium Shipments

Small quantities of oxides were shipped out as container “heels” and larger amounts were shipped out without being processed in the feed plant. UF<sub>6</sub> was shipped out as RU to ORGDP and Portsmouth. Shipments are summarized in Table 3.3-1

**Table 3.3-1, Recycled Uranium Shipments**

<b>Total Oxides Shipped</b>	<b>UF<sub>6</sub> To ORGDP</b>	<b>UF<sub>6</sub> To Portsmouth</b>
1420.5 MTU	620 MTU	793 MTU

### 3.4 Recycle Uranium Waste

Ash from UF<sub>6</sub> feed plant operations, vacuum dust, and filter cake solids comprised the largest portion of waste produced in the production of UF<sub>6</sub> from recycle oxides and U recovery operations. U recovery and recovery of material from ash continued until 1976. In 1976, after shutdown of the feed plant, most of this waste was shipped to FMPC. This material is classified as recycle waste because of its assay and source in plant operations.

**Table 3.4-1, Recycled Uranium Waste Shipped**

<b>Material Type</b>	<b>Total MTU Shipped</b>
Tower Ash	68.7
U <sub>3</sub> O <sub>8</sub>	23.2
Filter Cake Solids	2
Incinerator Ash	23.4
Vacuum Dust	225
Misc. Solids	25
Impure UF <sub>4</sub>	130
Misc.	1

Current inventory indicates that 18 containers of waste are still on site containing 850g Np, 3g Pu, and 50g <sup>99</sup>Tc. This material is stored in 55-gallon drums or 85-gallon over-packs in C-746-Q. This material contains greater than 100 nCi TRU per g.

### 3.5 Recycle Uranium Scrap

No recycle uranium scrap, per sec, has been identified at PGDP. Items in this category are identified as waste in Section 3.4, Table 3.4-1. Most of the RU waste has been shipped to FMPC. However, 149 kg of code 78 material (ash from feed plant for recycle) is currently maintained in inventory per plant accountability records. This material could be considered scrap, but it has value and was originally intended to be recycled.

An additional quantity of recycle uranium as UF<sub>6</sub> (1267 MTU) is maintained in inventory at PGDP per plant accountability records and could also be categorized as scrap (See Table 3.6-1).

### 3.6 Inventory as of March 31, 1999

A survey of NMC&A documents indicates the following materials produced from recycle uranium are still present on site.

**Table 3.6-1, Recycled Uranium (UF<sub>6</sub>) at PGDP**

<b>Material Type</b>	<b>Code 19</b>	<b>Code 24</b>	<b>Code 30</b>
Metric Tons U	3.5 (heels)	335	929
Number of Cylinders	104	53	125
<b>Total MTU</b>			<b>1267.5</b>

It should be noted that the code 30 material consists of RU from European and Asian commercial reactors. This material was reprocessed in Western Europe prior to shipment to the United States. Prior to June 1988, the DOE policy was to accept commercial RU and give full feed and separative work unit (SWU) content credit against their SWU purchases. In June 1988 DOE provided a revised policy to their customers which stipulated three things: 1) U had to come from

fuel originally produced by DOE, 2) DOE would provide RU product to those giving RU feed and NU (natural) product to those giving natural feed, 3) RU feed must meet DOE specifications. In addition a surcharge was to be imposed to cover the additional costs associated with RU. Once this policy came out the receipt of RU stopped.

From 1969 and 1988 over 1500 MTU of commercial RU was delivered to DOE. Originally these deliveries occurred at ORGDP. More than 500 MTU of material was fed to ORGDP between 1969 and 1985. Once ORGDP was shutdown the inventory at ORGDP and new RU inventory was sent to Paducah. By this time, additional concerns by the customers led to the decision to not feed the RU to the cascade. However, the cylinders still had to be returned to the customer. Many of the cylinders were 2.5 ton with some 10 and 14-ton cylinders as well. Paducah was directed to transfer the material from customer owned cylinders to DOE owned cylinders. The last deliveries occurred in August of 1988. PGDP finished transferring the material a year later in 1989. Although none of the material was fed to the cascade, during the process of transferring and heeling the cylinders, about 7 MTUs of the RU material was evacuated to the cascade.

The DOE conducted several studies in the late 1980's and 1990's to see how to capitalize on the then growing RU market. Different scenarios were looked at as far as dedicating a plant to feed only RU, campaigning RU, etc., and calculating the surcharges required. Around 1992, DOE decided that due to the increasing emphasis on health, safety and environmental issues, RU enrichment was determined to be a nonviable option. In addition, the RU market did not materialize to the extent once thought probable (Throbridge Report).

#### **4.0 Constituents in Recycled Uranium**

This section presents the range of concentrations of the TRU (Pu and Np) and <sup>99</sup>Tc constituents in the RU processed by the site. Available data for reporting these concentrations is somewhat limited. Assumptions used and available data sources are described in the following sections. All concentration units are on a uranium basis unless otherwise noted.

##### **4.1 Analytical Laboratories**

The PGDP Analytical Laboratories are located in building C-710. The laboratories have been in operation at the site since plant startup and were comprised of the ASTM, Industrial Hygiene, Infra-Red Spectrometry, Emission Spectrometry, Sampling, Uranium Analysis, Trouble Shooting, Quality Control, Radiochemistry, Metallurgy, Mass Spectrometry, Counting Preparation, Alpha and Beta Counting, and Fission Training Laboratories.

Since the initial staffing of the laboratory, technicians have undergone formal training to ensure appropriate qualification and use of appropriate laboratory techniques. Documented training modules have supported the training and qualification process. Additionally, the laboratory has had controlled procedures to ensure the quality of operations. A laboratory quality assurance program has been in place since the early phases of plant operation. Quality control standards that are indistinguishable from regular plant samples have been sent through the analysis process periodically along with plant samples and data accumulated continuously. From these data, control charts have been plotted, and precision and accuracy of a particular group of samples determined. Applicable QC data (believed to be in the vault) concerning the constituents in recycle uranium has not been accessed for RU transfers from the PGDP site due to problems with information assessability.

#### 4.2 Analytical Results for Plutonium in Uranium Materials Shipped

Analytical data for Pu in materials shipped from the Paducah facility are extremely limited. Some data can be inferred based on the fact that some of the shipments made were never processed in the Plant and the same reasoning (incoming results/assumptions) can thus be used in determining an approximate Pu content. This will be applied to oxides shipped back to the producers.

##### 4.2.1 Plutonium Specification in Recycled Uranium

UO<sub>3</sub> received from Hanford from the UO<sub>3</sub> plant has historically had a Pu specification of 10 ppb on a U basis. This specification was in effect at the startup of the UO<sub>3</sub> plant at Hanford and when the first production quantity U shipments were made in 1952 to ORGDP (Hanford Recycled Uranium Project Presentation Outline, dated 2/24/00). This specification is assumed to be applicable to all shipments of reactor tails UO<sub>3</sub> regardless of source.

##### 4.2.2 Impurity Concentrations In RU Shipped (Plutonium)

More significant quantities of Pu were present in a concentrated form in the feed plant ash and incinerator ash. Because of the chemical actions of Pu most of it will concentrate in the ash. Several interplant shipments of ash were made in order to reclaim Np and U from the ash. Large shipments of ash, along with other materials associated with feed plant closure, were made to FMPC in 1976. A final shipment of ash was made to FMPC in FY-80. Ash shipments are tabulated in Table 4.2-1.

**Table 4.2-1, Feed Plant Ash Shipments, MTU**

<b>FY</b>	<b>To ORGDP MTU</b>	<b>To NLO MTU</b>	<b>To ORNL MTU</b>	<b>To Y-12 MTU</b>
53				
54				
55				
56				35.2
57			3.8	1
58			17.8	
59	2.2		2.5	
60				
61				
62				
63	1.6			
64				
65				
66				
67				
68				
69				
70				
71				
72				
73				



<b>FY</b>	<b>To ORGDP MTU</b>	<b>To NLO MTU</b>	<b>To ORNL MTU</b>	<b>To Y-12 MTU</b>
<b>74</b>				
<b>75</b>				
<b>76</b>		36.7		
<b>77</b>		0.3		
<b>78</b>				
<b>79</b>				
<b>80</b>		23.4		
<b>81</b>				
<b>82</b>				
<b>Totals</b>	<b>3.8</b>	<b>60.4</b>	<b>24.1</b>	<b>36.2</b>

Several types of materials shipped to FMPC as part of the feed plant closure in FY-76 were also sources of TRUs shipped. These are listed in table 4.2-2 along with the limited data available on Pu concentrations noted.

**Table 4.2-2, Miscellaneous to FMPC from Feed Plant Closure**

	<b>FY</b>	<b>Material</b>	<b>Av.Pu Con., ppb</b>	<b>Total MTU</b>	<b>Pu, G</b>
<b>1</b>	<b>76*</b>	Feed Plant Ash	35.2	36.6	1.3
<b>2</b>	<b>76*</b>	Vacuum Dust	21.1	241	
<b>3</b>	<b>76</b>	Filter Cake	Not available	1	
<b>4</b>	<b>76</b>	Incinerator Ash		19.9	
<b>5</b>	<b>77</b>	Filter Cake		2.2	
<b>6</b>	<b>77</b>	Incinerator Ash		.6	
		Items 2-6			18.8
	<b>80**</b>	Feed Plant Ash	37 to 3118 ppb	23.4	14.2

\* Data from letter from C. C. Hopkins to Mr. C. R. Chapman dated October 21, 1975

\*\*Totals calculated from results of 16 hoppers analyzed by FMPC.

Inferences can also be drawn as to the amount of Pu shipped to other sites as RU in the form of UF<sub>6</sub>. Only a very small amount of Pu present in the oxides and transferred to the cylinder was subsequently vaporized to the cascade (< 0.1%) and Pu mobility within the cascade was very limited (based on analyses of cascade dusts). Thus, the amount of Pu present in the product UF<sub>6</sub> is expected to be below detection limits. This is confirmed by actual analyses. A large number of analyses were made on product and tails cylinders since 1972. Data from 1973-1981 are not available on a yearly basis but were summarized in a letter of March 19, 1992 from Mr. C. R. Beverly to Mr. J. C. Hodges. This data indicate 60 product and tails cylinders were analyzed in that time frame with only two product cylinders having a positive result for Pu. One cylinder was at 0.02 ppb and the other 0.06 ppb. Report KY/L-1239 identifies the result as suspect to contamination during the extraction, and that Pu probably was not present.

Additional data from FY-81 to FY-97 extracted from fiscal year summaries of analyses of product tails UF<sub>6</sub> are presented in Table 4.2-3.

**Table 4.2-3, Pu Concentrations Noted in Paducah Product and Tails Cylinders**

<b>FY</b>	<b>Type</b>	<b>No. of Cylinders</b>	<b>ppb Pu</b>
<b>81</b>	Pad. Prod	6	<0.01
	Pad. Tails	3	<0.01
<b>82</b>	Pad. Prod	NA.	NA.
	Pad. Tails	NA.	NA.
<b>83</b>	Pad. Prod	12	<0.01
	Pad. Tails	13	<0.01
<b>84</b>	Pad. Prod	13	<0.01
	Pad. Tails	12	<0.01
<b>85</b>	Pad. Prod	10	<0.01
	Pad. Tails	4	<0.01
<b>86</b>	Pad. Prod	13	<0.01
	Pad. Tails	12	<0.01
<b>87</b>	Pad. Prod	15	<0.01
	Pad. Tails	13	<0.01
<b>88</b>	Pad. Prod	13	<0.01
	Pad. Tails	12	<0.01
<b>89</b>	Pad. Prod	12	<0.01
	Pad. Tails	15	<0.01
<b>90</b>	Pad. Prod	12	<0.01
	Pad. Tails	13	<0.01
<b>91</b>	Pad. Prod	12	<0.01
	Pad. Tails	20	<0.01
<b>92 &amp; 93</b>	Pad. Prod	31	<0.01
	Pad. Tails	17	<0.01
<b>94</b>	Pad. Prod	12	<0.01
	Pad. Tails	7	<0.01
<b>95</b>	Pad. Prod	14	<0.01
	Pad. Tails	9	<0.01
<b>96</b>	Pad. Prod	12	<0.01
	Pad. Tails	10	<0.01
<b>97</b>	Pad. Prod	15	<0.01
	Pad. Tails	26	<0.01

#### **4.3 Analytical Results for Neptunium in Uranium Materials Shipped**

Early data on Np concentration are extremely limited. Data referenced in a letter of March 19, 1992 from Mr. C. R. Beverly to Mr. J. C. Hodges indicate that a total of 62 product and 40 tails cylinders were analyzed in the period from 1974-1982. All tails cylinders contained < 5 ppb Np/g U and only a few product cylinders (including 1 at 25 ppb, whose validity has not been verified) were above the reporting level. (Note: this report gives levels of Np in both dpm /g U and ug/ g U. There is a conversion or typographical error since 8 dpm is equivalent to 5 nanograms. The correct concentration for Np should therefore be in ng/g U or ppb.)

##### **4.3.1 Neptunium Specification in Recycled Uranium**

Prior to 1966 there was a UF<sub>6</sub> specification of 150 TRU alpha dpm/g U. This translates into 0.1ppm Np or less assuming no other TRUs are present (K/ETO-30 dated September 1990). In

1966, the standard for UF<sub>6</sub> was changed to 1500 alpha dpm/gmU (i.e., 1ppm Np or less). However, no specific specification is known to exist for Np in reactor tails in the form of UO<sub>3</sub>. As noted previously, Np measurements were not made at PGDP on incoming tails prior to FY-57 and only limited analytical Np data has been found on any reactor tails receipts after that date. Summary data from the Smith report (KY/L-1239 appears to be the best source information.

#### 4.3.2 Impurity (Neptunium) Concentrations in Recycled Uranium Shipped

Data amassed from fiscal year summaries of UF<sub>6</sub> analyses available on product and tails Np concentrations are presented in table 4.3-1.

**Table 4.3-1, Neptunium Concentrations in Paducah Product and Tails Cylinders Sampled**

<b>FY</b>	<b>TYPE</b>	<b>No. of Cylinders</b>	<b>ppb Np</b>
<b>81</b>	Pad. Prod	6	< 1
	Pad. Tails	3	< 1
<b>82</b>	Pad. Prod		
	Pad. Tails		
<b>83</b>	Pad. Prod	12	< 5
	Pad. Tails	13	< 5
<b>84</b>	Pad. Prod	13	< 5
	Pad. Tails	12	< 5
<b>85</b>	Pad. Prod	10	< 5
	Pad. Tails	4	< 5
<b>86</b>	Pad. Prod	13	< 5
	Pad. Tails	12	< 5
<b>87</b>	Pad. Prod	15	< 5
	Pad. Tails	13	< 5
<b>88</b>	Pad. Prod	13	< 5
	Pad. Tails	12	< 5
<b>89</b>	Pad. Prod	12	< 5
	Pad. Tails	15	< 5
<b>90</b>	Pad. Prod	12	< 5
	Pad. Tails	13	< 5
<b>91</b>	Pad. Prod	12	< 5
	Pad. Tails	20	< 5
<b>92 &amp; 93</b>	Pad. Prod	31	< 5
	Pad. Tails	17	< 5
<b>94</b>	Pad. Prod	12	< 5
	Pad. Tails	7	< 5
<b>95</b>	Pad. Prod	14	3.5
	Pad. Tails	9	< 5
<b>96</b>	Pad. Prod	12	2
	Pad. Tails	10	3
<b>97</b>	Pad. Prod	15	2.8
	Pad. Tails	26	3
<b>98</b>	Pad. Prod		
	Pad. Tails		

Other materials containing Np were shipped to FMPC such as feed plant ash, vacuum dust, and filter cake solids for recovery of U present. Much of the material has no analytical data associated with it with exceptions of ash, vacuum dust, and filter cake. Attempts are being made to estimate total amounts from FMPC data on feed material blended from these miscellaneous materials. The total amounts of Np shipped are summarized in table 4.3-2

**Table 4.3-2, Neptunium in Materials Shipped to FMPC**

<b>FY</b>	<b>Material</b>	<b>Av. Np, ppm</b>	<b>Total MTU</b>	<b>Np, g</b>
<b>76*</b>	Feed Plant Ash	0.89	36.6	32.5
<b>76*</b>	Vacuum Dust	0.33	241	79.5
<b>76</b>	Filter Cake	Not available	1	30
<b>76</b>	Incinerator Ash		19.9	21.7
<b>77</b>	Filter Cake		2.2.	4
<b>77</b>	Incinerator Ash		.6	1
<b>80**</b>	Feed Plant Ash	1.330-24.85	23.4	173

\* Data from letter from C. C. Hopkins to Mr. C. R. Chapman dated October 21, 1975

\*\* Totals calculated from results of 16 hoppers analyzed by FMPC.

#### **4.4 Analytical Results for Technetium in Uranium Materials Shipped**

Data abstracted from KY/L-1239, Appendix 10 yields  $^{99}\text{Tc}$  values for the period of highest measured concentration of  $^{99}\text{Tc}$  in product shipments. Prior to the installation of a  $\text{MgF}_2$  trap in C-310, analysis for  $^{99}\text{Tc}$  in Paducah product averaged 3.2ppm. After the installation of the trap, the measured average dropped to 0.15ppm for a period of about four months. No routine measurements were made on  $^{99}\text{Tc}$  in product  $\text{UF}_6$  from 1963 until FY-72. Traps were dumped in 4 to 5 month intervals.  $^{99}\text{Tc}$  was recovered for a time from the  $\text{MgF}_2$  traps and shipped to ORNL. When interest in recovering  $^{99}\text{Tc}$  from the traps waned, trap dumping become lax, resulting in  $^{99}\text{Tc}$  again appearing in the product. Routine measurements for  $^{99}\text{Tc}$  in the product cylinder were initiated in FY-72. Typical data from the period FY-72 thru FY-81 are shown in table 4.4-1 and include cylinder concentration ranges.

##### **4.4.1 Technetium Specification in Recycled Uranium**

$^{99}\text{Tc}$  content for RU was only indirectly controlled by the specification for  $\text{UF}_6$  on beta activity due to FP at 10% that for of aged natural uranium. If geometry factors and shielding were taken into account 8 of 10ppm  $^{99}\text{Tc}$  would be required to equal the specification level. A  $^{99}\text{Tc}$  threshold of 0.4ppm/U basis was proposed in 1982 but never implemented. As noted in section 4.7.1, no specification for incoming  $\text{UO}_3$  was ever established and consequently  $\text{UO}_3$  shipments from PGDP to other sites had no such specification.

**Table 4.4-1,  $^{99}\text{Tc}$  Concentration in Paducah Product Cylinders (from KY/L-1239)**

<b>Fiscal Year</b>	<b>Type</b>	<b>Cylinders</b>	<b>Ppm <math>^{99}\text{Tc}</math></b>	<b>Range</b>
<b>72</b>	Product	5	0.67	0.19 to 1.7
<b>73</b>	Product	13	4.5	<0.1 to 20
<b>74</b>	Product	12	6.1	<0.2 to 20
<b>75</b>	Product	12	1	<0.2 to 3
<b>76</b>	Product	27	0.98	0.03 to 3.7
<b>76A &amp; 77</b>	Product	35	.071	0.02 to 6.3

<b>Fiscal Year</b>	<b>Type</b>	<b>Cylinders</b>	<b>Ppm <sup>99</sup>Tc</b>	<b>Range</b>
<b>78</b>	Product	20	0.19	0.02 to 0.97
<b>79</b>	Product	24	0.14	<0.01 to 0.40
<b>80</b>	Product	26	0.09	<0.01 to 0.38
<b>81</b>	Product	16	0.025	<0.01 to 0.18

#### **4.4.2 Impurity Concentrations In Recycled Uranium Shipped (Technetium)**

Analytical results for <sup>99</sup>Tc in Paducah product and tails cylinder filled in FY-72 thru FY-97 are shown in Table 4.4-2. These data are derived from “Summary of Fiscal Years UF<sub>6</sub> Analysis Reports” and from a letter of March 19, 1992, from Mr. C. R. Beverly to Mr. J. C. Hodges.

**Table 4.4-2, Concentration of <sup>99</sup>Tc in Paducah Product and Tails Cylinders**

<b>Fiscal Year</b>	<b>Type</b>	<b>Cylinders</b>	<b>ppm <sup>99</sup>Tc</b>
<b>72</b>	Paducah Tails	*	<0.01
	Paducah Product	5	0.67
<b>73</b>	Paducah Tails	*	<0.01
	Paducah Product	13	4.5
<b>74</b>	Paducah Tails	*	<0.01
	Paducah Product	12	6.1
<b>75</b>	Paducah Tails	*	<0.01
	Paducah Product	12	1.0
<b>76</b>	Paducah Tails	*	<0.01
	Paducah Product	27	0.98
<b>77</b>	Paducah Tails	*	<0.01
	Paducah Product	35	0.71
<b>78</b>	Paducah Tails	*	<0.01
	Paducah Product	20	0.19
<b>79</b>	Paducah Tails	*	<0.01
	Paducah Product	24	0.14
<b>80</b>	Paducah Tails	*	<0.01
	Paducah Product	26	0.09
<b>81</b>	Paducah Tails	3	<0.01
	Paducah Product	16	0.025
<b>82</b>	Paducah Tails	*	<0.01
	Paducah Product	22	<0.01
<b>83</b>	Paducah Tails	13	<0.01
	Paducah Product	22	<0.01
<b>84</b>	Paducah Tails	12	<0.01
	Paducah Product	31	<0.01
<b>85</b>	Paducah Tails	4	<0.01
	Paducah Product	33	<0.01
<b>86</b>	Paducah Tails	12	<0.01
	Paducah Product	26	<0.01
<b>87</b>	Paducah Tails	13	<0.01
	Paducah Product	15	<0.01
<b>88</b>	Paducah Tails	12	<0.01
	Paducah Product	13	<0.01

<b>Fiscal Year</b>	<b>Type</b>	<b>Cylinders</b>	<b>ppm <sup>99</sup>Tc</b>
<b>89</b>	Paducah Tails	15	<0.01
	Paducah Product	23	<0.01
<b>90</b>	Paducah Tails	13	<0.01
	Paducah Product	12	<0.01
<b>91</b>	Paducah Tails	20	<0.01
	Paducah Product	12	<0.01
<b>92 &amp; 93</b>	Paducah Tails	17(2)**	<0.01-(0.01)
	Paducah Product	31(14)**	<0.01-(0.01)
<b>94</b>	Paducah Tails	7	<0.01
	Paducah Product	30	0.009
<b>95</b>	Paducah Tails	9	<0.005
	Paducah Product	28	0.0376
<b>96</b>	Paducah Tails	20	<0.01
	Paducah Product	29	<0.01
<b>97</b>	Paducah Tails	26	<0.01
	Paducah Product	36	<0.01

\* Data for tails material were not broken down by fiscal year. All analyses were less than 0.01ppm and total cylinders analyzed were 37.

\*\*The values in parentheses indicate results at the detectable limit of 0.01ppm/U basis. Variations noted in the “less than” values are due to changes made in the measurement procedure as methods changed for the analysis.

Additional shipments of <sup>99</sup>Tc or <sup>99</sup>Tc -bearing material were made to other sites. A total of 620 MTU UF<sub>6</sub> prepared from recycle oxides were shipped to ORGDP (an assumed concentration of 7ppm <sup>99</sup>Tc for oxides and 85% transferred from cylinders yields 3.6 kg <sup>99</sup>Tc) and 793 MTU UF<sub>6</sub> reactor tails shipped to Portsmouth (same assumption yields 4.6 kg <sup>99</sup>Tc). Return of 1768 MTU (code 1496 enriched) of unprocessed oxides to producers at a concentration of 7 to 16ppm <sup>99</sup>Tc accounts for a further 26 kg of <sup>99</sup>Tc. Approximately 25 kg of <sup>99</sup>Tc was transferred to ORNL in solutions for recovery of the <sup>99</sup>Tc. Additional large quantities of <sup>99</sup>Tc have been shipped out as an impurity in Paducah product. Calculations of this quantity have not been finalized, but initial data examination reveal that shipments prior to FY-75 transferred 79 kg to Portsmouth and 117 kg to ORGDP as enriched Paducah product. After FY-75 an additional 1kg of <sup>99</sup>Tc was transferred to ORGDP and 5kg <sup>99</sup>Tc to PORTS in Paducah product. Conversations with analysts confirmed the validity of assuming a <sup>99</sup>Tc concentration of ½the < values reported. Approximately 1 kg <sup>99</sup>Tc was shipped out in waste. Total <sup>99</sup>Tc transfers enumerated are 262 kg.

Other small shipments of spent trapping materials such as alumina, magnesium fluoride, and miscellaneous solid materials and solutions were made to producers (chiefly FMPC) but <sup>99</sup>Tc content and total mass has not been determined.

#### **4.5 Analytical Results for Plutonium in Uranium Materials Received**

Reactor tails materials in the form of UO<sub>3</sub> were received at the PGDP from both Hanford and/or Savannah River from FY-1953 through FY-1975. Limited Pu analysis data by both Paducah and Hanford on UO<sub>3</sub> carload composites from Hanford have been identified starting in November 1953(KYL-58) with the first shipment from Hanford identified to date being received in September of 1953. These analytical data have not been correlated with specific receipts due to the inability to associate specific carload composite samples with receipts. Limited comparative

Pu analytical results reported by Paducah and Hanford for some subsequent receipts of Hanford reactor tails are also available but are similarly limited.

The first receipts identified from Savannah River were received in January of 1955(KY-110). Summary data on UO<sub>3</sub> received from Savannah River showing Paducah and Savannah River comparative Pu analytical results have also been identified in KY-62, Part 3, issued in April of 1955. It is assumed that the analytical data in KY-62, Part 3 is associated with the shipments received in January 1955 but as noted, these data have not been correlated with specific receipts. The PGDP and Savannah River analytical results are shown in Table 4.5-1 and are included for comparative purposes. The PGDP and Hanford analytical results are shown in Table 4.5-2 and are also included for comparative purposes. In general the PGDP results are higher than those for Savannah River. Smith pointed out that the Paducah data were biased high in this time period due to other alpha emitting impurities, e.g., Np not known to be present. Source documents are also included in Table 4.5-1 and 4.5-2

**Table 4.5-1, Savannah River UO<sub>3</sub> Received, MTU  
(Measured Concentrations of Plutonium)**

<b>Lot No.</b>	<b>Date</b>	<b>PPB Pu SR</b>	<b>PPB Pu PGDP</b>	<b>~ % U – 235</b>	<b>Doc No., KYL</b>
<b>4</b>	1955	2.5	3	0.688	62-1
<b>5</b>	1955	6.1	7	0.684	62-1
<b>6</b>	1955	6.1	12	0.683	62-1
<b>7</b>	1955	4	11	0.684	62-1
<b>8</b>	1955	2.2	11	0.684	62-1
<b>9</b>	1955	1.7	7	0.684	62-1
<b>10</b>	1955	2.8	10	0.684	62-1
<b>11</b>	1955	5.5	8	0.684	62-1
<b>12</b>	1955	4.4	9	0.684	62-1
<b>13</b>	1955	2.7	4	0.684	62-1
<b>14</b>	1955	2.5	3	0.683	62-2
<b>15</b>	1955	2	10	0.693	62-2
<b>16</b>	1955	2.3	10	0.683	62-2
<b>17</b>	1955	1.7	7	0.679	62-2
<b>18</b>	1955	1.8	5	0.680	62-2
<b>19</b>	1955	1.5	8	0.680	62-3
<b>20</b>	1955	1.5	4	0.680	62-3
<b>21</b>	1955	1.4	7	0.680	62-3
<b>22</b>	1955	1.5	5	0.680	62-3
<b>23</b>	1955	2.7	6	0.680	62-3
<b>24</b>	1955	2.5	5	0.680	62-3
<b>41</b>	1955	2	4	0.680	62-4
<b>42</b>	1955	<1	4	0.680	62-4
<b>43</b>	1955	1	5	0.681	62-4
<b>44</b>	1955	<2	5	0.683	62-4
<b>45</b>	1955	1	4	0.684	62-4
<b>46</b>	1955	1	3	0.683	62-5
<b>47</b>	1955	1	5	0.684	62-5
<b>48</b>	1955	3	6	0.685	62-5
<b>49</b>	1955	4.1	3.9	0.685	62-5
<b>50</b>	1955	1	3	0.686	62-5

Lot No.	Date	PPB Pu SR	PPB Pu PGDP	~ % U – 235	Doc No., KYL
51	1955	1	3	0.686	62-6
52	1955	2	7	0.686	62-6
53	1955	8	13	0.686	62-6
54	1955	7	14	0.685	62-6
68	1955	7	3	0.674	62-6
69	1955	6	2	0.674	62-7
70	1955	3	1	0.674	62-7
71	1955	4	1	0.674	62-7
95	1955	5	<2	0.674	62-7
151-155	1962		4	0.608	269-1
157-159	1962		1	0.609	269-1
160-162	1962		1	0.609	269-1
163-165	1962		<1	0.614	269-1
178-180	1962		2	0.611	269-1
193-195	1962		2	0.603	269-1
208-210	1962		1	.06	269-2

\*Mean: Savannah River= 3.4 ppb (SR analysis), 6ppb (PGDP analysis)

The limited analytical data on shipments received from Savannah River never exceeded the 10ppm specification according the shippers analysis and only infrequently, per the PGDP analysis (See Table 4.5-1) which were considered biased high by Smith. In addition, Table 4.5-2 summarizes limited Pu analytical data by both Paducah and Hanford for Hanford carload composites samples (Hanford data agrees well with Paducah data).

**Table 4.5-2, Plutonium in Hanford UO<sub>3</sub> Received Feed Plant Carload Composite Samples**

Lot	Date (CY)	Hanford	Paducah	~%235	Doc # KYL
104	1953	<5	1	.647	KYL-28
106	1953	<5	1	.672	KYL-28
108	1953	<5	1	.659	KYL-28
110	1953	<5	1	.673	KYL-28
112	1953	<5	1	.680	KYL-28
114	1953	6	1	.673	KYL-28
115	1953	<5	1	.666	KYL-28
117	1953	<5	1	.669	KYL-28
118	1953	<5	<1	.667	KYL-28
120	1953	<5	<1	.669	KYL-28
122	1953	<5	<1	.673	KYL-28 Part 2
124	1953	<5	1	.669	KYL-28 Part 2
126	1953	<5	1	.667	KYL-28 Part 2
127	1953	<5	3	.664	KYL-28 Part 2
128	1953	<5	1	.667	KYL-28 Part 2
129	1953	<5	1	.668	KYL-28 Part 3
130	1953	<5	1	.661	KYL-28 Part 3
131	1953	<5	<1	.667	KYL-28 Part 3
132	1953	<5	2	.682	KYL-28 Part 3
133	1953	<5	1	.685	KYL-28 Part 3
134	1954	<5	2	.676	KYL-28 Part 4



Lot	Date (CY)	Hanford	Paducah	~%235	Doc # KYL
135	1954	7	2	.683	KYL-28 Part 4
136	1954	6	1	.681	KYL-28 Part 4
137	1954	10	3	.672	KYL-28 Part 4
138	1954	<5	2	.66 to .68	KYL-28 Part 4
139	1954	<5	1	.656	KYL-28 Part 5
140	1954	19	21	.655	KYL-28 Part 5
141	1954	10	13	.654	KYL-28 Part 5
142	1954	6	3	.656	KYL-28 Part 5
143	1954	5	2	.653	KYL-28 Part 5
144	1954	6	5	.651	KYL-28 Part 5
146	1954	6	4	.669	KYL-28 Part 5
147	1954	<5	<1	.675	KYL-28 Part 5
148	1954	13	9	.670	KYL-28 Part 5
149	1954	12	13	.667	KYL-28 Part 5
150	1954	5	4	.671	KYL-28 Part 7
151	1954	<5	2	.664	KYL-28 Part 7
152	1954	6	2	.661	KYL-28 Part 7
153	1954	<5	1	.661	KYL-28 Part 7
154	1954	<5	1	.669	KYL-28 Part 7
155	1954	5	1	.664	KYL-28 Part 8
156	1954	<5	1	.658	KYL-28 Part 8
157	1954	<5	1	.654	KYL-28 Part 8
158	1954	<5	1	.653	KYL-28 Part 8
159	1954	<5	1	.661	KYL-28 Part 8
166	1954	<5	<1	.659	KYL-28 Part 10
167	1954	<5	<1	.657	KYL-28 Part 10
168	1954	<5	<1	.661	KYL-28 Part 10
171	1954	<5	<1	.666	KYL-28 Part 10
172	1954	<5	<1	.658	KYL-28 Part 10
170	1954	<5	1	.666	KYL-28 Part 10
176	1954	<5	1	.660	KYL-28 Part 10
177	1954	<5	5	.662	KYL-28 Part 10
179	1954	<5	5	.662	KYL-28 Part 10
181	1954	<5	1	.656	KYL-28 Part 10
182	1954	<5	1	.662	KYL-28 Part 10
184	1954	<5	1	.674	KYL-28 Part 10
185	1954	5	1	.658	KYL-28 Part 10
188	1954	<5	1	.668	KYL-28 Part 10
198	1954	<5	1	.665	KYL-28 Part 15
200	1954	<5	1	.660	KYL-28 Part 15
202	1954	<5	2	.654	KYL-28 Part 15
204	1954	<5	2	.655	KYL-28 Part 15
206	1954	<5	4	.668	KYL-28 Part 16
210	1954	5	1	.669	KYL-28 Part 16
213	1954	<5	1	.660	KYL-28 Part 16
214	1954	<5	2	.665	KYL-28 Part 16
216	1954	<5	1	.667	KYL-28 Part 16
218	1954	<5	2	.667	KYL-28 Part 16
219	1954	<5	1	.653	KYL-28 Part 17
220	1954	<5	1	.654	KYL-28 Part 17

Lot	Date (CY)	Hanford	Paducah	~%235	Doc # KYL
222	1954	<5	1	.653	KYL-28 Part 17
224	1954	<5	2	.660	KYL-28 Part 17
226	1954	3	2	.649	KYL-28 Part 17
UA-200	1958	3	2	.652	KYL-28 Part 19
UA-220	1959	3	2	.650	KYL-166 Part 2
UA-240	1959	8	7	.644	KYL-166 Part 2
UA-260	1959	7	7	.646	KYL-166 Part 4
UA-280	1959	2	4	.649	KYL-166 Part 9
306	1959		4	.630	KYL-166 Part 11
310	1959		5	.627	KYL-166 Part 11

\*Mean: Hanford FY-54, 3.3 ppb (assuming  $\frac{1}{2}$  reporting limit), PGDP analysis = 3.3 ppb.  
1958-1959, Hanford 5ppb, Paducah 4.8ppb

#### 4.5.1 Plutonium Specification in Recycled Uranium

UO<sub>3</sub> received from Hanford from the UO<sub>3</sub> plant has historically had a Pu specification of 10 ppb on a U basis (See Savannah River and Hanford Reports). This specification was in effect at the startup of the UO<sub>3</sub> plant at Hanford and when the first production quantity U shipments were made in 1952 to ORGDP (Hanford Recycled Uranium Project Presentation Outline, dated 2/24/00). This specification is assumed to be applicable to all shipments reactor tails UO<sub>3</sub> regardless of source.

#### 4.5.2 Impurity (Plutonium) Concentrations in Incoming Recycled Uranium

Smith (KY/L-1239, dated March 1984) summarized the quantities and concentrations of Pu received in reactor tails at PGDP, based upon amount of material fed to the cascade as material codes 524(SRT), 624(HRT) and enriched reactor returns (ERT) This summary is shown in Table 4.5-3 which shows concentrations and their ranges for material received by source. The total amount of Pu received was estimated at 328 grams. These data (Table 4.5-3) are considered the best initial estimate of Pu receipts for the PGDP. Data examined in the course of the preparation of this report have supported the validity of this initial estimate within 10% (300g vs. 328g). A different approach was used in the examination of source types (oxides and UF<sub>6</sub> by originator), and independent validation of concentrations of Pu in the oxides. (See table 4.5-4)

**Table 4.5-3, Plutonium Received in Reactor Tails, UO<sub>3</sub>, (per Smith Report)**

Stream	Quantity of Reactor Tails, MTU	Concentration Ppb, Pu, U Basis	Range Ppb PU, U Basis	Received Grams
HRT and SRT before FY-67	74,898	4*	2 to 6	272
HRT after FY-67	22,236	2.2	0.9 to 3.6	45
SRT after FY-67	1,890	1.0	0.2 to 2.5	2
ERT	2,154	4.5	3.1 to 9.0	9
			<b>Total</b>	<b>328</b>

\*Results were biased high since pulse height analyzer was not available to check extractions for alpha emitting impurities; however, the magnitude of bias is not known.

**Table 4.5-4, Receipts Pu from UO<sub>3</sub> in Grams by FY and Source (calculated in this work)**

<b>FY</b>	<b>ppb Pu SR</b>	<b>Pu-SR g</b>	<b>ppb Pu -Han</b>	<b>Pu-Han g</b>	<b>ppb Pu -NLO</b>	<b>Pu-NLO g</b>	<b>misc. Pu g</b>
53							0.3
54			2.51	5.4			0.6
55	4.8	1.4	1.72	4.6			
56	3.3	0.6	1.98	7.9			
57	2.3	1.3	2.51	13.6			
58	3.5	0.8	3.25	19.8			
59			5.69	29.1			
60			3.04	15.7			
61	4	0.3	3.97	23.8			
62	1.4	2.2	4, 4.5	25.5			
63	1.4	1.9	4.76	27.5			
64	1.6	2.6	5.42	22.6			
65							
66							
67			4	57.7			
68							
69	1	2.8	2.2	7.8			
70	1	0.6	2.2				
71			2.2, 4.5	1.8	2.2, 4.5	2.8	
72			2.2, 4.5	9.9	2.2, 4.5	4.1	
73			2.2, 4.5	2.0	2.2, 4.5	0.8	
74							
75							
76							
77						2.2	
<b>Subtotal g</b>		<b>14</b>		<b>275</b>		<b>10.0</b>	<b>0.9</b>
<b>Total g</b>							<b>300</b>

\*Where possible, data on Pu concentrations were derived from calculations made using analytical data derived from KY-L-411. Average concentrations specified by the Smith report were used to fill in data gaps, primarily in FY-62 and after FY-65.

Plutonium estimates on oxide receipts were also performed using average constituent data developed by FMPC with the following results:

Savannah River - 9,231 MTU @ 2.81ppb = 25.9g Pu  
 Hanford - 74,479 MTU @ 2.59ppb = 192.9g Pu  
 FMPC - 3,929 MTU @ 2.59ppb = 10.2g Pu  
 Harshaw & ORGDP 265 MTU @ 2.59ppb = 0.7g Pu

Total estimated Pu results thus indicated are approximately 230g. Our calculated result lies between this estimate and the original estimate of Smith of 328g Pu received.

#### **4.6 Analytical Results for Neptunium in Uranium Materials Received**

Smith (KY/L-1239, dated March 1984) summarized the quantities and concentrations of Np received in reactor tails at PGDP. This summary is shown in Table 4.6-1, which shows concentrations and their ranges for different materials received. Smith noted that Np

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measurements were not made on reactor tails received prior to FY-57 and that those made after that time were limited. Analyses were performed on monthly composite samples of UO<sub>3</sub> after receipt. The total amount of Np received was estimated at 18.4 kg (See also Table 5.3-1). These data are considered the best initial estimate of Np receipts for the PGDP.

**Table 4.6-1, Neptunium Received in Reactor Tails, (per Smith Report)**

Stream	Quantity of Reactor Tails Tons U	Average Concentration ppm Np U Basis	Concentration Range, ppm Np U Basis	Apparent No. Received Kg
HRT and SRT before FY-67	74,898	0.24	0.01 to 0.6	16.3
HRT after FY-67	22,326	0.09	0.05 to 0.27	1.8
SRT after FY-67	1,890	0.12	<0.01 to 0.22	0.2-
ERT	2,154	0.05	0.01 to 0.11	0.01
			<b>Total</b>	<b>18.4</b>

Fed to Cascade: 18.4 x 25% = 4.6 kg.

Calculations were also made on total net receipts of Np from all oxide sources using the above referenced concentrations. Table 4.6-2 lists those calculations and amount of Np. The differential is primarily due to consideration of other sources in addition to those from Savannah River and Hanford sites. Concentrations noted are a combination of values from the Smith report and those calculated from KY-L-411.

**Table 4.6-2, Neptunium Receipts, (calculated in this work)**

FY	S.R. UO <sub>3</sub> , MTU	Np conc, ppm	Total Np g	Han UO <sub>3</sub> , MTU	Np conc, ppm	Np Total g	Misc. UO <sub>3</sub> , MTU	Np conc, ppm	Np g
53							68	0.24	16.32
54				2170	0.24	520.8	152	0.24	36.48
55	283	0.24	67.92	2647	0.24	635.3			
56	171	0.24	41.04	3972	0.24	953.3	0.2	0.24	0.048
57	565	0.24	135.6	5428	0.61	3311			
58	229	0.013	2.977	6104	0.62	3784			
59				5112	0.12	613.4	21.1	0.24	5.064
60				5151	0.1	515.1			
61	81	0.013	1.053	6001	0.08	480.1	10.2	0.24	2.448
62	1546	0.06	92.76	5630	0.43	2421	0.3	0.24	0.072
63	1344	0.11	147.8	5775	0.18	1040	0.3	0.24	0.072
64	1603	0.02	32.06	4178	0.12	501.4			
65									
66									
67				14422	0.24	3461			
68									
69	2811	0.12	337.3	3537	0.24	848.9	0.2	0.24	0.048
70	597	0.12	71.64				13.6	0.24	3.264
71	34	0.12	4.08	722	0.24	173.3	761	0.24	182.64
72	-34	0.12	-4.08	3072	0.24	737.3	1779	0.24	426.96
73				556	0.24	133.4	386	0.24	92.64

FY	S.R. UO <sub>3</sub> , MTU	Np conc, ppm	Total Np g	Han UO <sub>3</sub> , MTU	Np conc, ppm	Np Total g	Misc. UO <sub>3</sub> , MTU	Np conc, ppm	Np g
74									
75									
76									
77							1002	0.24	240.48
<b>S Tot.</b>			930.2			20129			1006.5
<b>Total</b>									<b>22066</b>

#### 4.6.1 Neptunium Specification in Recycled Uranium

Prior to 1966 there was a UF<sub>6</sub> specification of 150 TRU alpha dpm/gmU. This translates into 0.1ppm Np or less assuming no other TRUs are present (K/ETO-30 dated September 1990). In 1966, the standard for UF<sub>6</sub> was changed to 1500 alpha dpm/gmU (i.e., 1ppm Np or less). However, no specific specification is known to exist for Np in reactor tails in the form of UO<sub>3</sub>. As noted above, Np measurements were not made at PGDP on incoming tails prior to FY-57 and limited analytical Np data (only summary data from Smith and KY-L-411) has been found on reactor tails receipts.

#### 4.6.2 Impurity (Neptunium) Concentrations in Incoming Recycled Uranium

From Table 4.6-1 it can be seen that the Np content of reactor tails varies according to the source of material, the type of material, and the date received. The 1967 transition date shown in Tables 4.6-1 and 4.6-2 represents a process change in the production of UO<sub>3</sub> from the Redox to the Purex process. Enriched reactor tails with a U assay greater than 0.711 percent U-235 contributed only in a minor way to the total Np received. Raw analytical data supporting the Smith results for Np have not been identified.

#### 4.7 Analytical Results for Technetium in Uranium Materials Received

The best source of information concerning the concentration of <sup>99</sup>Tc in reactor tails identified to date is also from the Smith report (KY/L-1239, dated March 1984). This information is shown below in Table 4.7-1. The total amount of <sup>99</sup>Tc received at PGDP is estimated at 661 kg. Concentrations and their ranges are included. Raw analytical data to support the Smith results have not been identified.

**Table 4.7-1, Technetium Received in Reactor Tails (per Smith Report)**

Stream	Reactor Tails in MTU	Ave. Concentration ppm <sup>99</sup> Tc, U Basis	Concentration Range ppm <sup>99</sup> Tc, U Basis	Apparent <sup>99</sup> Tc Received in Kg
Hanford Reactor Tails (HRT) and Savannah River Reactor Tails (SRT)	90104	7	4-10	630
Enriched Reactor Tails (ERT)	1958	16	11-27	31
			<b>Total</b>	<b>661 kg</b>

**Table 4.7-2,  $^{99}\text{Tc}$  Received at Paducah by Source (calculated in this work)**

<b>Source</b>	<b>MTU</b>	<b>Concentration</b>	<b>Total Kgs <math>^{99}\text{Tc}</math></b>
Savannah oxides	9230	7ppm/ U basis	64.6
Hanford oxides (depleted)	71770	7ppm/ U basis	502.4
Hanford oxides (enriched)	2707	16ppm/ U basis	43.3
FMPC oxides (depleted code 528)	387	7ppm/ U basis	2.7
FMPC oxides (enriched)	594	16ppm/ U basis	9.5
RU $\text{UF}_6$ (ORGDP & PORTS)	10668	5.8ppm/ U basis	61.9
Foreign $\text{UF}_6$	1542		
<b>Total</b>	<b>96898</b>	<b>Not applicable</b>	<b>684.4</b>

Table 4.7-2 provides a separate estimate of the total mass of  $^{99}\text{Tc}$  received by material type but uses the average concentrations assumed by Smith. No independent verification of the concentration of  $^{99}\text{Tc}$  has been identified. The 5.8ppm  $^{99}\text{Tc}$  concentration for the RU  $\text{UF}_6$  in Table 4.7-2 has been corrected from the 7ppm used by Smith to account for  $^{99}\text{Tc}$  not transferred to the  $\text{UF}_6$  Clyinder. The data agreement with that estimated by Smith is very good, if consideration is given that his data represents MTU of material fed to the cascade, and the total of  $^{99}\text{Tc}$  above includes approximately 10 kgs of  $^{99}\text{Tc}$  that were eventually shipped out in RU oxides without being fed.

#### **4.7.1 Technetium Specification in Recycled Uranium**

No specification relative to the  $^{99}\text{Tc}$  content of incoming reactor tails has been identified. In 1974 the specification for  $^{99}\text{Tc}$  in feed  $\text{UF}_6$  was only indirectly controlled by the specification on beta activity due to FP (KY-708, Rev. 2.). This specification states that the maximum beta activity due to FP will be no more than ten percent of the beta activity of aged natural U. This in effect limits the  $^{99}\text{Tc}$  value in feed  $\text{UF}_6$  to approximately 10ppm on a U basis (KY-L-708, Rev. 2). Assuming that 100% of the  $^{99}\text{Tc}$  present in the reactor tails was converted to a volatile species in the feed plant, this specification would in effect establish a 10ppm limit for reactor tails  $\text{UF}_6$ . Based on Table 4.7-1 and using a weighted average of reactor tails processed at the PGDP feed plant, this 10ppm limit would in general have been met by incoming reactor tails.

#### **4.7.2 Impurity (Technetium) Concentrations in Incoming Recycled Uranium**

From Table 4.7-1 it can be seen that the  $^{99}\text{Tc}$  concentration in reactor tails received varies according to the type of tails material received. ERT (enriched reactor tails, received from Hanford) are tails materials with a U-235 assay greater than 0.711% and typically contain about two times the  $^{99}\text{Tc}$  concentration found in tails with a U assay of less than 0.711 %. Only relatively small quantities of ERT material were received and constituted only about two percent of the total tails material received. The amount of  $^{99}\text{Tc}$  received from ERT materials was about four percent of the total  $^{99}\text{Tc}$  received.

## 5.0 Mass Balance Activities

### 5.1 Annual Mass Balances of Recycled Uranium

An annual mass balance of RU at the Paducah plant has already been presented in section 3.2 and 3.3 of this report, and Table 3.2-1 enumerates total flow by type for both shipments and receipts. A summary of that data is repeated for convenience in Table 5.1-1. Mass Flow of constituent summary is depicted in Figure 5.1-2 Figure 5.1-1 depicts the RU mass flow by source and material type for the PGDP

**Table 5.1-1, Summary of Recycled Uranium Mass Flow at Paducah, MTU**

	<b>Receipts</b>			<b>Shipments</b>		
<b>FY</b>	<b>MTU UO<sub>3</sub></b>	<b>MTU UF<sub>6</sub></b>	<b>MTU UF<sub>4</sub>*</b>	<b>MTU UO<sub>3</sub></b>	<b>MTU UF<sub>6</sub></b>	<b>MTU UF<sub>4</sub>*</b>
<b>Total</b>	88192	12829.4	495	1444	1412.9	407.3
<b>Net UO<sub>3</sub></b>						86748
<b>Net UF<sub>6</sub></b>						11416.5
<b>Net UF<sub>4</sub> *</b>						87.7

\*UF<sub>4</sub> with assay (%<sup>235</sup>U) less than 0.68 and greater than 0.72 and assumed to be from reactor returns.

Figure 5.1-1, Recycled Uranium Mass Flow

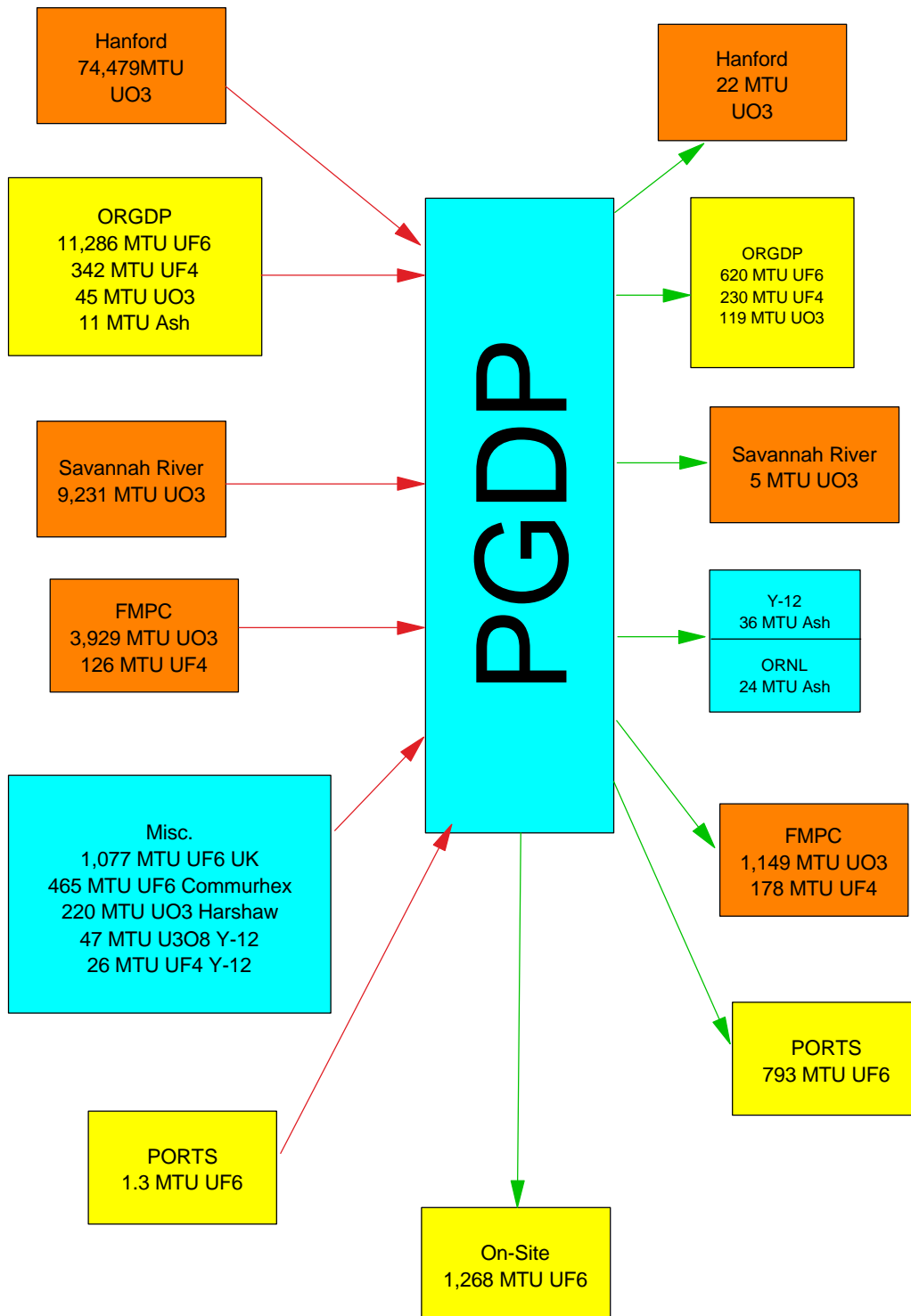
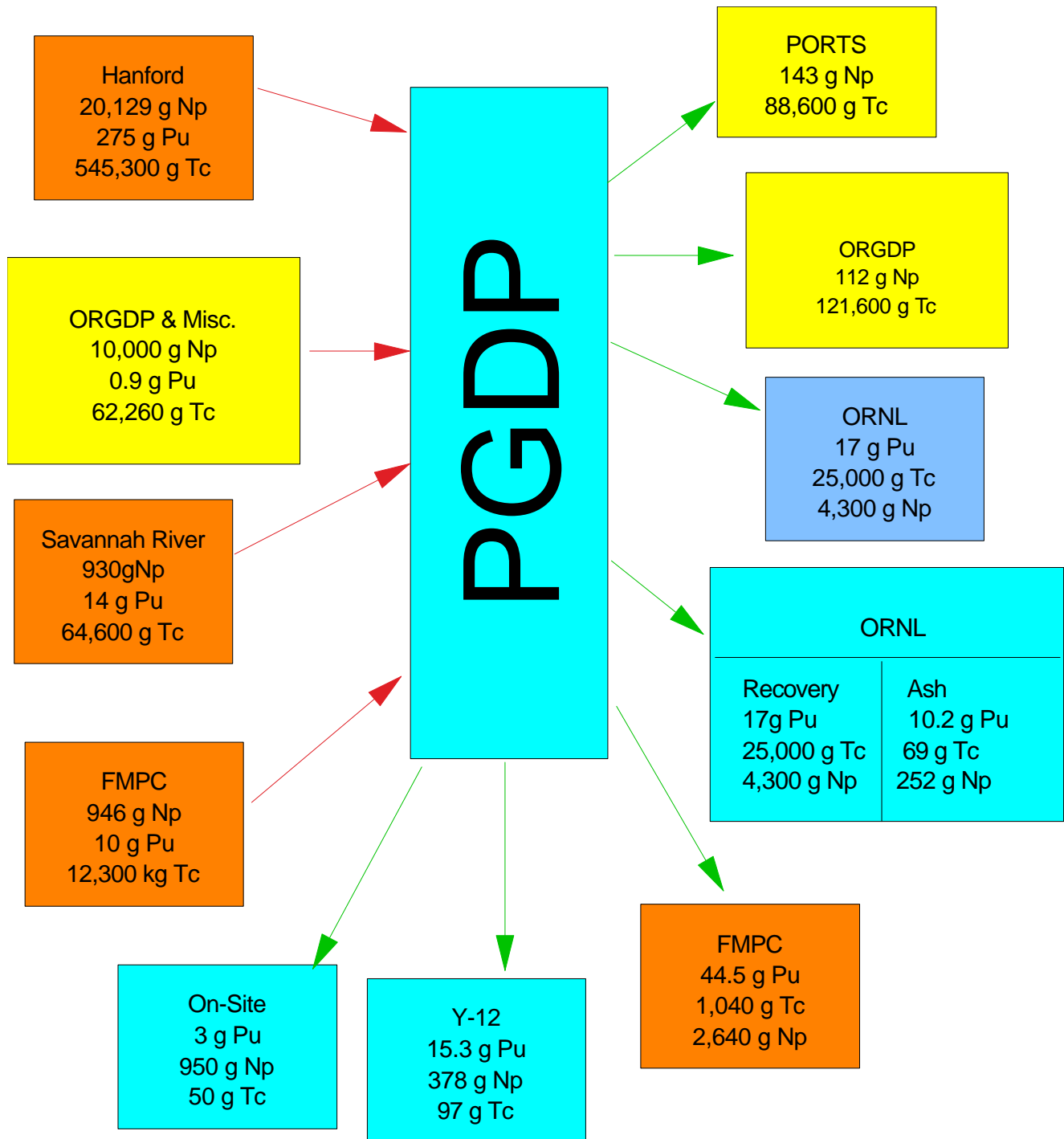




Figure 5.1-2, Constituent Mass Flow



## 5.2 Annual Mass Balances of Plutonium in Recycled Uranium.

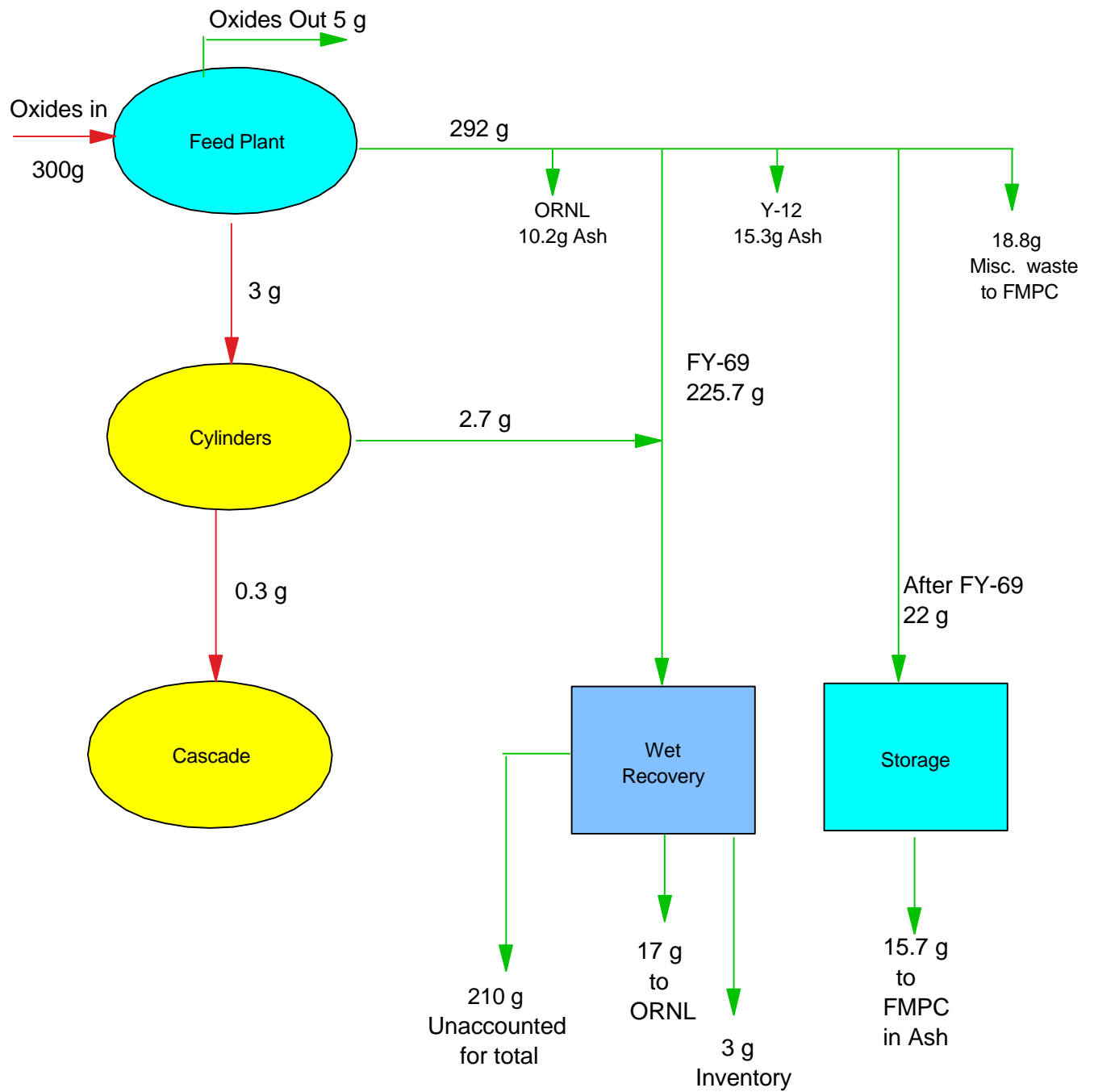
Reactor tail material as  $\text{UO}_3$  was received from Hanford, Savannah River and FMPC containing Pu at various levels dependent on the type of process used to separate the U from reactor tails (i.e. REDOX or PUREX extraction methods), and the assay of the material (depleted or enriched). Good data on Pu concentration are somewhat sketchy but average concentration based on data from the Smith report (KY/L-1239) can be used to achieve an expected mass of Pu received from these sources. The receipts are presented in Table 5.2-1. Figure 5.2-1 depicts the plutonium mass flow into and out of PGDP.

**Table 5.2-1, Receipts Pu from  $\text{UO}_3$  in Grams by Fiscal Year and Source**

<b>FY</b>	<b>ppb Pu SR</b>	<b>Pu-SR g</b>	<b>ppb Pu -Han</b>	<b>Pu-Han g</b>	<b>ppb Pu -NLO</b>	<b>Pu-NLO g</b>	<b>misc. Pu g</b>
<b>53</b>							0.3
<b>54</b>			2.51	5.4			0.6
<b>55</b>	4.8	1.4	1.72	4.6			
<b>56</b>	3.3	0.6	1.98	7.9			
<b>57</b>	2.3	1.3	2.51	13.6			
<b>58</b>	3.5	0.8	3.25	19.8			
<b>59</b>			5.69	29.1			
<b>60</b>			3.04	15.7			
<b>61</b>	4	0.3	3.97	23.8			
<b>62</b>	1.4	2.2	4, 4.5	25.5			
<b>63</b>	1.4	1.9	4.76	27.5			
<b>64</b>	1.6	2.6	5.42	22.6			
<b>65</b>							
<b>66</b>							
<b>67</b>			4	57.7			
<b>68</b>							
<b>69</b>	1	2.8	2.2	7.8			
<b>70</b>	1	0.6	2.2				
<b>71</b>			2.2, 4.5	1.8	2.2, 4.5	2.8	
<b>72</b>			2.2, 4.5	9.9	2.2, 4.5	4.1	
<b>73</b>			2.2, 4.5	2.0	2.2, 4.5	0.8	
<b>74</b>							
<b>75</b>							
<b>76</b>							
<b>77</b>						2.2	
<b>Subtotal g</b>		<b>14</b>		<b>275</b>		<b>10.0</b>	<b>0.9</b>
<b>Total g</b>							<b>300</b>

\* Where possible, data on Pu concentrations were derived from calculations made using analytical data derived from KY-L-411. Average concentrations specified by the Smith report were used to fill in data gaps, primarily FY-62 and after FY-65.

Figure 5.2-1, Pu Mass Flow



### 5.3 Annual Mass Balances of Neptunium in Recycled Uranium

Np-containing reactor tails material, in the form of UO<sub>3</sub>, was received at the Paducah plant site from both Hanford and Savannah River from FY-1953 through FY-1975. The presence of Np in this material was not recognized until 1956. Prior to that time, the Np content of the reactor tails is very uncertain which has led to problems in estimating the quantity of Np received during these early years. No reactor tails material has been fed to the Paducah cascade since September 11, 1975.

A flow chart showing the Np Mass Balance Flow is shown in Figure 5.3-1, Np Mass Flow. A summary of all feed streams to the Paducah cascade during this time period is presented in Table 5.3-1, where both the cascade feed prepared from reactor tails material and the total cascade feed are shown for each year in terms of tons of U. While the percentage of feed material made from reactor tails varied widely from year to year (as high as 65% in FY-1973), these percentage variations were largely the result of variations in the other feed materials; the feed rate of reactor tails material was actually fairly constant over most of the period, i.e., between 6,000 and 10,000 tons U per year, with smaller quantities being fed in the early years of the program

It is estimated that 23 kg of Np was received at Paducah. The estimated quantities of Np (in kilograms) received yearly at Paducah in the reactor tails material is shown in Table 5.3-1 (on both a yearly and cumulative basis) for FY-1953 through FY-1976.

It is assumed that the 7.3 MTU foreign RU introduced to the cascade in 1986 through 1989 essentially contributed no Np to the PGDP site. Refer to section 3.6 for additional discussion on this material that was received in the material form of UF<sub>6</sub>.

In addition to the Np received in reactor tails material, some Np was returned from the Oak Ridge and Portsmouth sites during the CIP/CUP, associated with scrap metal (principally barrier) removed from the respective cascades and sent to Paducah for smelting and metal recovery operations. However, the quantity of Np associated with this scrap was relatively small, probably amounting to, at most, a few tenths of a kg of Np. Because of the large uncertainty associated with the quantity of Np received in the reactor tails material, as discussed above, this small additional Np input to the Paducah site has been neglected in the material balance.

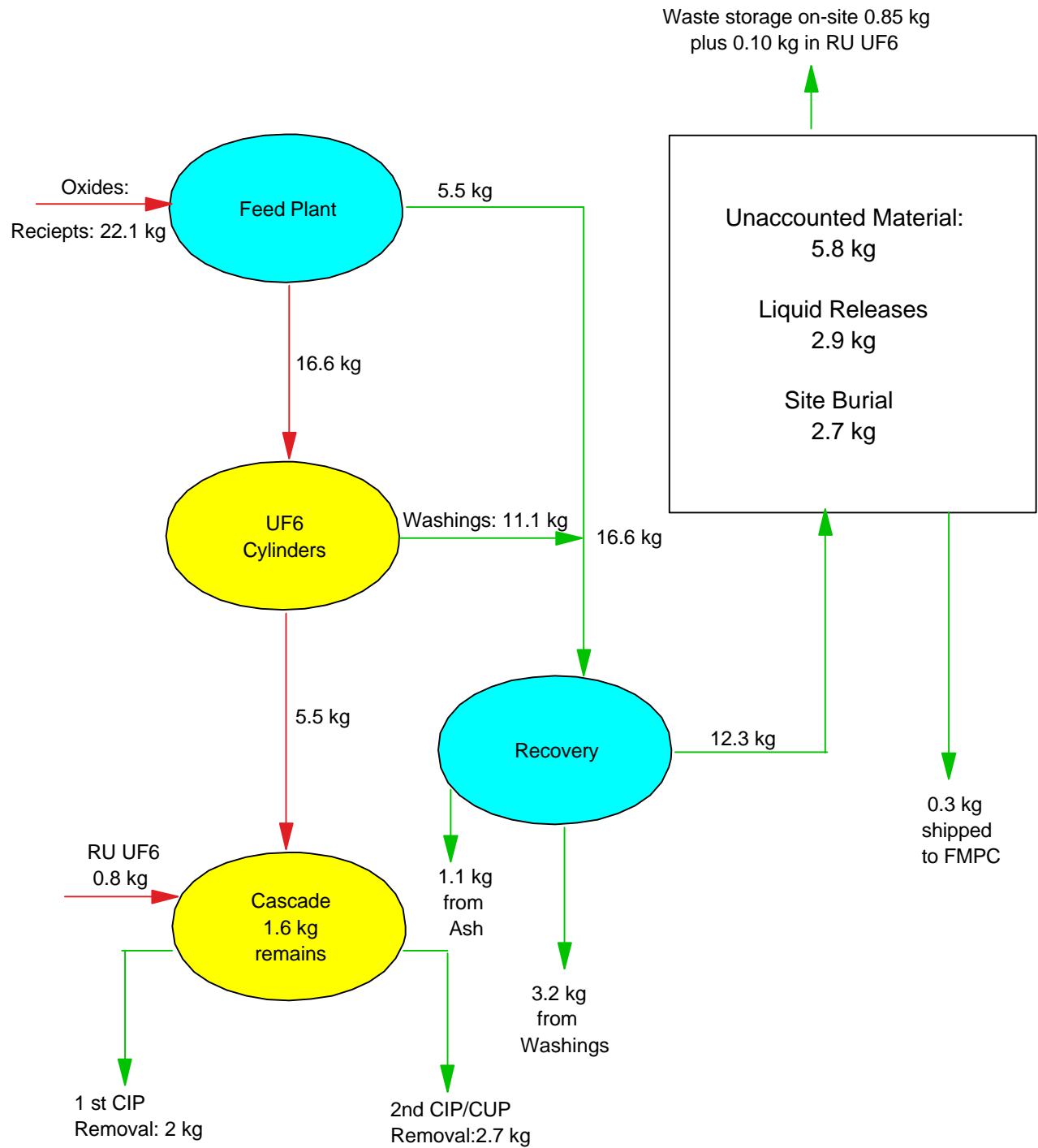
**Table 5.3-1, Neptunium Receipts and Cascade Feed at PGDP<sup>3</sup>**

FY	S.R. UO <sub>3</sub> , MTU	Np conc, ppm	Total Np g	Han UO <sub>3</sub> , MTU	Np conc, ppm	Np Total g	Misc. UO <sub>3</sub> , MTU	Np conc, ppm	Np g
53							68	0.24	16.32
54				2170	0.24	520.8	152	0.24	36.48
55	283	0.24	67.92	2647	0.24	635.3			
56	171	0.24	41.04	3972	0.24	953.3	0.2	0.24	0.048
57	565	0.24	135.6	5428	0.61	3311			
58	229	0.013	2.977	6104	0.62	3784			
59				5112	0.12	613.4	21.1	0.24	5.064
60				5151	0.1	515.1			
61	81	0.013	1.053	6001	0.08	480.1	10.2	0.24	2.448
62	1546	0.06	92.76	5630	0.43	2421	0.3	0.24	0.072

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FY	S.R. UO3, MTU	Np conc, ppm	Total Np g	Han UO3, MTU	Np conc, ppm	Np Total g	Misc. UO3, MTU	Np conc, ppm	Np g
63	1344	0.11	147.8	5775	0.18	1040	0.3	0.24	0.072
64	1603	0.02	32.06	4178	0.12	501.4			
65									
66									
67				14422	0.24	3461			
68									
69	2811	0.12	337.3	3537	0.24	848.9	0.2	0.24	0.048
70	597	0.12	71.64				13.6	0.24	3.264
71	34	0.12	4.08	722	0.24	173.3	761	0.24	182.64
72	-34	0.12	-4.08	3072	0.24	737.3	1779	0.24	426.96
73				556	0.24	133.4	386	0.24	92.64
74									
75									
76									
77							1002	0.24	240.48
<b>S Tot.</b>			930.2			20129			1006.5
<b>Total</b>									<b>22066</b>

Figure 5.4-1 depicts the technetium mass flow to and from the PGDP.

**Figure 5.3-1, Np Mass Flow**

#### 5.4 Annual Mass Balances of Technetium in Recycled Uranium

<sup>99</sup>Tc estimates for material were derived using the following assumptions:

- A. <sup>99</sup>Tc concentration in recycled oxides was 7ppm/ U basis for depleted oxides and 16-ppm/ U basis for enriched oxides.
- B. Approximately 95% of the <sup>99</sup>Tc was transferred to the UF<sub>6</sub> cylinder.
- C. Since <sup>99</sup>Tc analysis data were unavailable for initial years of operation, <sup>99</sup>Tc estimates in product were made based on the percentage of product for that year from RU
- D. Although <sup>99</sup>Tc has a lag time in residence in the cascade before reaching the product withdrawal point, for total estimates of <sup>99</sup>Tc in product, it was assumed to immediately transfer and end when recycle feed ended.
- E. When <sup>99</sup>Tc analysis data were available, after FY-72, it was used to calculate <sup>99</sup>Tc content in product.

Annual estimates of <sup>99</sup>Tc in RU are presented in table 5.4-1

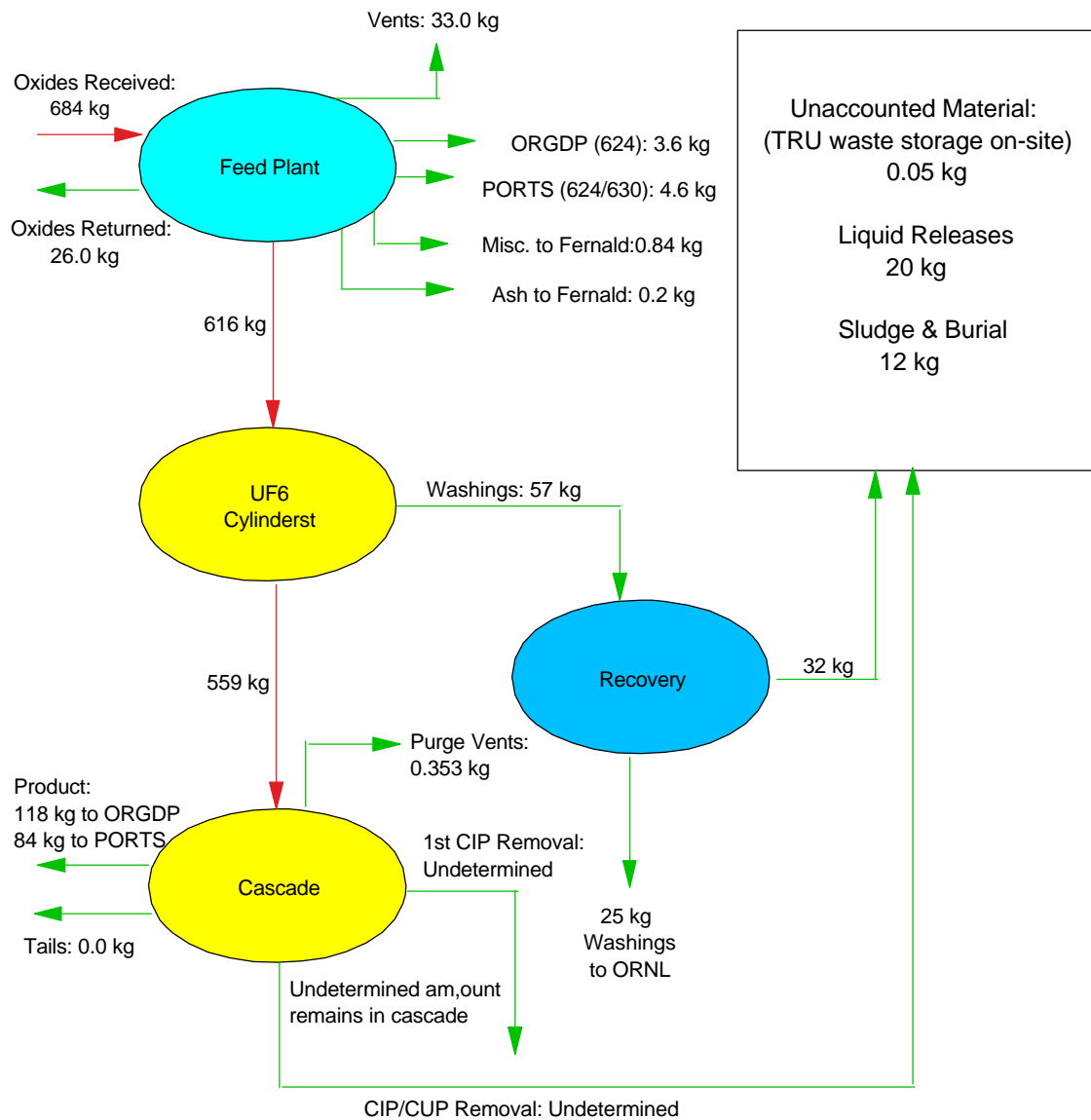
**Table 5.4-1, Annual Estimates of <sup>99</sup>Tc in Recycled Uranium, kg**

<b>FY</b>	<b><sup>99</sup>Tc-oxides In</b>	<b><sup>99</sup>Tc-RU UF<sub>6</sub> In</b>	<b><sup>99</sup>Tc- RU UF<sub>6</sub> Out</b>	<b><sup>99</sup>Tc in Prod. UF<sub>6</sub> Out</b>
<b>53</b>	0.5	6.1		1.3
<b>54</b>	16.3	9.2		3.4
<b>55</b>	20.5	3.8	0.7	1.7
<b>56</b>	29	14	0.2	4.5
<b>57</b>	42	16		14.4
<b>58</b>	44.3	4.9	0.1	9.5
<b>59</b>	35.9	2.8	0.4	5.3
<b>60</b>	36.1	3.2		5.6
<b>61</b>	42.6	0.4		4.9
<b>62</b>	58.5			4.8
<b>63</b>	49.8			3.2
<b>64</b>	40.4			2.2
<b>65</b>				
<b>66</b>				
<b>67</b>	101			
<b>68</b>				
<b>69</b>	24.5		3.18	7.8
<b>70</b>	4.3			9.2
<b>71</b>	15.6			
<b>72</b>	47.1			2.7
<b>73</b>	9.5			15.3
<b>74</b>				21.5
<b>75</b>				3.6
<b>76</b>				3.3
<b>76a &amp; 77</b>	7			2.1
<b>78</b>				0.5
<b>79</b>				0.4
<b>80</b>				0.5
<b>81</b>				0.2
<b>82</b>				0.1

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<b>FY</b>	<sup>99</sup> <b>Tc-oxides In</b>	<sup>99</sup> <b>Tc-RU UF<sub>6</sub> In</b>	<sup>99</sup> <b>Tc- RU UF<sub>6</sub> Out</b>	<sup>99</sup> <b>Tc in Prod. UF<sub>6</sub> Out</b>
<b>83</b>				0.07
<b>84</b>				0.03
<b>85</b>				0.05
<b>86</b>				0.02
<b>87</b>				0.04
<b>88</b>				0.04
<b>89</b>				0.04
<b>90</b>				0.04
<b>91</b>				0.05
<b>92</b>				0.04
<b>93</b>				0.05
<b>94</b>				0.04
<b>95</b>				0.04
<b>96</b>				0.04
<b>97</b>				0.04
<b>98</b>				0.04
<b>99</b>				0.03
<b>total</b>	624.9	60.4	4.58	128.7



**Figure 5.4-1, Tc Mass Flow**

## **5.5 Worker Exposure to Plutonium, Neptunium, and Technetium from Recycled Uranium**

Workers exposures are discussed in Section 2 of this Report.

## **5.6 Environmental contamination from Plutonium, Neptunium, and Technetium in Recycled Uranium.**

Over the years, solid wastes and recyclable metals were stored or disposed of in various locations including two landfills, four scrap yards, and three radioactive material disposal sites. In addition, there were a number of smaller holding areas and special disposal sites. A burn pit in the northwest corner of the site was used for combustible waste until 1967. The landfill used for early construction rubble north of the Plant continued in operation as the Plant came on line, and another landfill outside the fence southwest of the Plant (known as the C-746-K Landfill) was utilized for steam plant ash disposal and evolved into a general debris landfill. Although there were some early specifications limiting placement of radioactive material in the landfills, there is no record of sampling to determine if radioactive material was placed in the landfill. Furthermore, since records indicate that floor sweepings were disposed of in the landfills and spills of green salt and yellowcake were commonly found in several areas of the Plant, it could be assumed that radioactive materials potentially could have been sent to the sanitary landfills. In addition, waste materials (including radioactively contaminated materials) potentially could have been disposed of in various areas outside the Plant fenced boundary in what is now the West Kentucky Wildlife Management Area (WKWMA). Some of the materials disposed of outside the Plant boundary have been identified as radioactive by subsequent site surveys or investigations.

Scrap metals from C-340, the cascades, the feed plant, and the C-720 maintenance shop went to C-746-F (classified burial), C-746-E (contaminated material yard), C-746-C (clean materials), or unclassified burial yards all within the PGDP security fence. From the beginning of Plant operations, efforts were made to try to control the spread of contamination and to separate contaminated materials from other waste. Pyrophoric uranium metal shavings were disposed of in the C-749 burial ground from 1957 to 1977. In the 1950s, U powder scrap from C-340 was dumped into onsite pits. The primary radioactive waste disposal site was the original C400 holding pond, which was converted into a solid waste disposal area in 1957. By 1977, over 6 million pounds of depleted U had been put into drums and placed in this disposal area.

In 1978 and 1979, the estimated amount of low level radioactive waste disposed of on site was 330,690 pounds annually, but this declined significantly to 18,000 pounds per year in the 1980s (Phase II Investigation). An overriding assumption regarding the stability of the radioactive disposal sites was that the underlying clay layer would prevent contamination from leaching into the groundwater and traveling off site.

In the early 1980s, the Material Terminal Management (MTM) Department began addressing hazardous waste disposal practices by working with waste generators to ensure that waste streams would be in compliance with RCRA requirements and by implementing standard operating procedures for waste management. Concurrently, the MTM and the Environmental Control Departments worked with regulators to obtain permits for storage, treatment, and disposal facilities, including the C-400 gold dissolver precipitation system and C-410 neutralization pit. Legacy hazardous waste was brought to several locations, including the C-733 Hazardous Waste Storage Area, the C-746-R Waste Solvent Storage Area, and the C-746-Q Hazardous Waste Storage Area. However, the absence of sufficient characterization to ensure long-term storage and

compliance with waste acceptance criteria for disposal has led to existing hazardous waste storage problems and the need for significant recharacterization (Phase II Investigation).

### **Air and Water Emissions**

Radioactive air emissions began with startup operations in 1952 and have continued to present. Air emissions from the site were released from process stacks, diffuse and fugitive emission sources, accidental releases, and a limited number of planned/controlled releases. From 1959 to 1974, the air emission reports consisted of ambient air monitoring. Starting in mid-1960, continuous ambient air samples were taken at four locations at the perimeter fence and were analyzed for alpha and beta contamination to provide input for annual reports on ambient air concentrations. In 1961, four additional ambient continuous air samplers were installed one mile outside the perimeter fence, although actual stack monitoring of emissions did not occur until the mid-1970s.

From 1975 through 1990, annual discharges to the atmosphere were based on stack measurements and reported in annual emission reports. It has been estimated that from 1952 to 1983, 60,000 kg of U were released to the atmosphere, 75 percent of this prior to 1965 and most from C-410 and C-340. A number of accidental releases of UF<sub>6</sub> occurred (perhaps as many as 15), during which more than 50 pounds of UF<sub>6</sub> were released. Dust and fugitive emissions were generally not calculated for the site from 1952 to 1990. (Phase II ESH Site Investigation Reference Report)

Construction of the PGDP incorporated systems and strategies for disposing of liquid effluents from production and support operations. Liquid effluents were released in a number of ways, including via the sanitary sewage and storm water drainage systems. The C-615 sewage treatment plant was used from the beginning to treat sanitary and sink wastes from production buildings. Other effluents were discarded either in batches or through continuous feed into ditches, ponds, and streams, and were usually monitored prior to release into the Big and Little Bayou Creeks, ultimately reaching the Ohio River.

Liquid effluent discharge limits for radionuclides have always been controlled under the AEC and ERDA regulations and later DOE orders as maximum permissible concentrations (MPCs) or radiation concentration guides (RCGs) in water.

It is assumed that the most significant liquid effluent discharge source at the Plant site was from the C-400 decontamination building. Wastes from this source included TCE from degreasing operations, contaminated liquids from cleaning operations, and various contaminated raffinate solutions from U, Np, and <sup>99</sup>Tc recovery operations. Additionally it can be summarized that all isotopes at the site were present in various concentrations at this facility and in its liquid waste streams, including U, Np, Pu, Th, and <sup>99</sup>Tc.

In 1988, concerns over residential water quality led to sampling of residential wells north of the Plant. TCE, an industrial degreaser, and <sup>99</sup>Tc, a radionuclide fission by-product from nuclear fuel, were discovered in the wells. This discovery prompted the government to provide municipal water free of charge to all residences and businesses in an area bounded by the Ohio River to the north, by the DOE property to the south, by Metropolis Lake Road to the east, and by Bethel Church Road to the west. Under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), DOE and the EPA developed an Administrative Consent Order, effective November 23, 1988, that established a schedule to investigate and remediate offsite groundwater contamination. Phase I of the CERCLA review, conducted in 1989 and 1990, identified contaminants of concern and solid waste management units (SWMUs) that could have

contributed to offsite contamination, outlined the physical characteristics of the SWMUs, and described the risk of offsite contamination. Phase II of the CERCLA review, conducted in 1990 and 1991, further assessed the risk of offsite contamination, characterized SWMUs that could have contributed to offsite contamination, and identified migration pathways for contaminants.

## **6.0 Results and Conclusions**

### **6.1 Explanation of Flow Paths**

#### **6.1.1 Flow of RU into PGDP**

PGDP processed RU from initial startup in 1952 through fiscal year 1989 with the exception of fiscal years 1965 through 1968, 1971, and 1977 through 1985. Five primary sources have been identified for RU material entering the PGDP:

- Oxides from Hanford for conversion to  $\text{UF}_6$  at PGDP – 74,477 MTU
- Oxides,  $\text{UF}_6$  feed, and ash scrap from the ORGDP that originated from Hanford, Savannah River, or miscellaneous foreign and domestic sources – 11,576 metric tons of U (MTU – 1000 kilograms of U)
- Oxides from Savannah River for conversion to  $\text{UF}_6$  at PGDP – 9,230 MTU
- Oxides,  $\text{UF}_6$  feed,  $\text{UF}_4$  and scrap ash from FMPC – 4,084 MTU
- $\text{UF}_6$  feed from miscellaneous foreign and domestic sources – 2006 MTU

Note: These RU receipts total approximately 101,363 MTU.

#### **6.1.2 Potential Flow Pathways of RU within PGDP**

Once an RU stream entered PGDP, the RU had the potential to reach various facilities and equipment via pathways associated with:

- $\text{UF}_6$  feed preparation C-420 Building and C-411, and C-410 Building Complex
- Enrichment cascade buildings C-331, C-333, C-335, C-337, and C-310
- Tails withdrawal C-315 Building
- Product withdrawal C-310 Building
- Support operations (including decontamination, U recovery, and other ancillary support facilities) and not limited to C-400, C-409, C-340, C-710, and C-720 buildings.

The pathways associated with each of these groups of operations are described in the following sections.

##### **6.1.2.1 $\text{UF}_6$ Feed Preparation Pathways**

The process of converting RU oxide to  $\text{UF}_4$  and to  $\text{UF}_6$  involved the following pathways:

- Unpacking and feeding of oxide to feed production process
- Hydrogen reduction of  $\text{UO}_3$  to  $\text{UO}_2$
- Hydro-fluorination to  $\text{UF}_4$
- Flame tower fluorination to  $\text{UF}_6$ , including generation of ash and filter residue Bldg. C-410
- Venting to atmosphere Bldg. C-410
- Collecting ash for U recovery and ultimate disposal and cleaning or disposal of filters

- Removal of  $\text{MgF}_2$  trap sludge, not limited to C-410 and C-420 buildings
- Pulverization of ash and other recovery processes
- Recycling of U in ash through fluorination
- Analytical laboratory sampling
- Packing of waste ash for storage prior to shipping
- Transfer of  $\text{UF}_6$  to cylinder

#### **6.1.2.2 Enrichment Cascade Operations Pathways**

Enrichment cascade operations involved the following pathways:

- Withdrawing  $\text{UF}_6$  from cylinder and feeding into the cascade
- Accumulation in cylinder heels
- Inadvertent releases of  $\text{UF}_6$  within cascade buildings or from piping between cascade buildings
- Venting to the atmosphere Bldg. C-310
- Removal of  $\text{MgF}_2$  trap sludge Bldg. C-410
- Analytical laboratory sampling

#### **6.1.2.3 Tails Withdrawal Pathways Building C-315**

Tails withdrawal involved the following pathways:

- Withdrawal of tails from cascade into cylinders
- Releases from ruptured tails cylinders in storage
- Analytical laboratory sampling

#### **6.1.2.4 Product Withdrawal Pathways Building C-310**

Product withdrawal involved the following pathways:

- Withdrawal of product from cascade into cylinders
- Releases from “pig tail” connections or other during withdrawal
- Analytical laboratory sampling

#### **6.1.2.5 Support Operations Pathways C-400, C-720, and Process Bldgs**

Support operations involved the following pathways:

- Cleaning of cylinder heels (potentially involving feed, product, or tails cylinders)
- Decontamination of equipment associated with feed, cascade, and other operations
- Routine and emergency maintenance operations at ancillary support facilities
- U recovery from oils, cleaning solutions, and other wastes
- Effluent, sludge, and other wastes from decontamination processes
- Incineration of certain wastes
- Scrap from equipment
- Removal or drainage of sludge from waste ponds
- Analytical laboratory sampling

### 6.1.3 Shipments of RU to off-site locations from PGDP

RU shipments:

- UF<sub>6</sub> product containing <sup>99</sup>Tc and non-detectable quantities of Pu and Np shipped to ORGDP – 85,230 MTU
- UF<sub>6</sub> product containing <sup>99</sup>Tc and non-detectable quantities of Pu and Np shipped to Portsmouth Gaseous Diffusion Plant (PORTS) – 119,167 MTU
- Enriched feed plant UF<sub>6</sub>, UF<sub>4</sub>, oxides, and ash scrap to FMPC – 9,070 – MTU

### 6.2 Identification of processes or facilities that involved potential worker exposure to RU constituents

Processes or facilities that involved potential worker exposure to RU constituents coordinate closely with the pathways described for the flow of RU within PGDP described in Section 6.1.2. These processes and their associated locations are summarized in Table 6.2-1.

**Table 6.2-1, Processes and Facilities Involving Potential Worker Exposure**

Process	Facilities	RU Constituent Involvement	Comments
Unpacking, feeding of UO <sub>3</sub> to feed production, reduction of UO <sub>3</sub> to UO <sub>2</sub> , hydrofluorination to UF <sub>4</sub>	C-420	RU constituents of concern had not been concentrated	
Flame tower fluorination of UF <sub>4</sub> to UF <sub>6</sub> , including generation of ash and filter residue	C-410, C-420	Pu and Np and some <sup>99</sup> Tc were concentrated in ash and residue	
Venting to atmosphere from flame tower fluorination	C-410, C-420	Venting did not involve concentrated RU constituents	Vent filters, however, were a potential concentration point.
Collecting ash for U recovery and cleaning of tower filters	C-410, C-420	Pu and Np and some <sup>99</sup> Tc were concentrated in ash and residue	Potential for worker exposure to concentrated RU constituents
Pulverization of tower ash and other recovery processes	C-410, C-420	Pu and Np and some <sup>99</sup> Tc were concentrated in ash and residue	Potential for worker exposure to concentrated RU constituents
Recycling of U in ash through fluorination	C-410, C-420	Pu and Np and some <sup>99</sup> Tc were concentrated in ash and residue	Potential for worker exposure to concentrated RU constituents
Packing of waste ash for storage prior to shipping	C-410, C-420	Pu and Np and some <sup>99</sup> Tc were concentrated in ash and residue	Potential for worker exposure to concentrated RU constituents
Transfer of UF <sub>6</sub> to cylinder from fluorination process	C-410, C-420	Pu and Np and some <sup>99</sup> Tc became further concentrated in cylinder heels	
Withdrawing UF <sub>6</sub> from cylinder and feeding into the cascade	Cascade feed points (see Table 2.2-1)	Pu and Np and some <sup>99</sup> Tc were concentrated in cylinder heels. Np and smaller amounts of Pu exiting cylinder with UF <sub>6</sub> collected at feeder head leading to cascade and within	

Process	Facilities	RU Constituent Involvement	Comments
		cascade near feed point.	
Venting to atmosphere from cascade	Cascade purge locations; (see Table 2.2-1) C-331, C-333, C-335, C-337, C-310	As RU moved higher into cascade, <sup>99</sup> Tc became concentrated, but Pu and Np were much diminished. Venting at higher points in cascade involved concentrated <sup>99</sup> Tc.	Traps were used to remove <sup>99</sup> Tc before venting
CIP/CUP equipment removal and other equipment removal, maintenance, and decontamination	Cascade buildings (see Table 2.2-1) C-270 Scrap Yard Burial Grounds	Np and lesser amounts of Pu collected near cascade feed points	Removal, handling, and decontamination of equipment near feed points could have exposed workers to concentrations of the small percentages of Np and Pu that actually entered the cascade
Recovery of deposits from cascade following various upgrades at PGDP	Cascade buildings (see Table 2.2-1)	Deposits could involve Pu, Np, or <sup>99</sup> Tc, depending on location in cascade	Removal of deposits could have exposed workers to concentrations of RU constituents, depending on location in cascade
Inadvertent releases of UF <sub>6</sub> within cascade buildings or from piping between cascade buildings	Cascade buildings and associated piping	Releases at higher points in the cascade could have exposed workers to higher concentrations of <sup>99</sup> Tc	
Withdrawal of tails from cascade into cylinders	Tails withdrawal points (see Table 2.2-1) C-315	Analytical data (Sect. 5.0) indicates that the tails stream did not contain significant quantities of RU constituents	
Withdrawal of product from cascade into cylinders	Product withdrawal points (see Table 2.2-1)	Analytical data (Sect. 5.0) indicates that product stream did not contain significant quantities of RU constituents	
Cleaning of heels from cylinders (feed cylinders were cleaned at PGDP.)	C-400	Pu and Np collected in the cylinder heels	Potential for worker exposure to concentrated RU constituents in feed cylinders
Decontamination and processing of equipment associated with feed, cascade, and other operations	C-410, C-420, C-400, C-333, and C-337	Decontamination of cascade equipment located near feed points during periods of large quantities of RU feed would be of greater concern because of Pu and Np	Activities involving the decontamination of equipment associated with the other processes and locations on this list could have exposed workers to RU constituents
Handling of scrap metal from equipment	C-746-E and E-1, C-746-C1 Scrap Metal Yard	Scrap from cascade equipment located near feed points during periods of large quantities of RU feed would be of greater concern because of Pu and Np	Scrap metal was processed for decontamination at C-400 before it was sent to C-746 Scrap Metal Yard
U recovery from and/or processing of oils, cleaning solutions, and	C-420, C-410, and C-400	Oils, solutions, and wastes associated with processes that concentrated RU constituents would be of greater	These processes could have exposed workers to RU constituents

Process	Facilities	RU Constituent Involvement	Comments
other wastes		concern. U recovery via solvent extraction concentrated $^{99}\text{Tc}$ in the sludge and precipitates formed from treatment of raffinate.	
Removal, transfer, and/or storage of sludge from facility treating $^{99}\text{Tc}$ concentrated in sludge (C-400)	C-747-4 and C-746-P	$^{99}\text{Tc}$ became concentrated in sludge associated with U recovery. Sludge was subsequently processed in C-403 Neutralization Pit	Dredging, moving, and storage of sludge from C-747-A and C-746-P could have exposed workers to concentrations of $^{99}\text{Tc}$
Analytical laboratory sampling	Analytical laboratories	Collection and analysis of samples from feed production and cascade processes could expose workers to RU constituents	

**6.3 Identification and Evaluation of Processes or facilities that involved Environmental Contamination from Identification and Evaluation of Processes or facilities that involved worker exposure to recycled uranium constituents.**

Processes and/or facilities that involved potential environmental contamination by RU constituents also coordinate closely with the pathways described for the flow of RU within PGDP described in Section 6.1.2. These processes and their associated locations are summarized in Table 6.3-1.

**Table 6.3-1, Processes and Facilities Involving Potential Environmental Contamination**

Process	Facilities	RU Constituent Involvement	Comments
Venting to atmosphere from flame tower fluorination	C-410 and C-420	Venting did not involve concentrated RU constituents	
Venting to atmosphere from cascade	Cascade purge locations; (see Table 2.2-1) C-310	As RU moved higher into cascade, $^{99}\text{Tc}$ became concentrated, but Pu and Np were much diminished. Venting at higher points in cascade involved concentrated $^{99}\text{Tc}$ .	Traps were used to remove $^{99}\text{Tc}$ before venting
Inadvertent releases of $\text{UF}_6$ within cascade buildings or from piping between cascade buildings	Cascade buildings and associated piping	Releases at higher points in the cascade could have involved concentrations of $^{99}\text{Tc}$	
Releases from ruptured tails cylinders in storage	Cylinder storage yards	Analytical data indicates that the tails stream did not contain significant quantities of RU constituents	PGDP- None of significant or measurable consideration
Outdoor storage of scrap metal from cascade	C-746-E, C-746-E1, C-746-C1, C-400 and E-403 pit Scrap Metal Yard	Scrap from cascade equipment located near feed points during periods of large quantities of RU feed would be of greater concern because of Pu and Np	Scrap metal was processed for decontamination at C-420 before it was sent to C-746 Scrap Metal Yard
Discharges of sludge from facility treating $^{99}\text{Tc}$ concentrated in sludge (C-400)	North South Diversion Ditch	Discharges from U recovery processes included waste streams with concentrations of $^{99}\text{Tc}$	



#### **6.4 Discussion of data sources**

The project team searched a variety of data collections and libraries at C-710, C-100, C-302 and C-304 locations to identify and retrieve data. Major data sources consulted and analyzed included:

- Nuclear Materials Control and Accountability (NMC&A) data, including shipping, receiving, and inventory records.
- Nuclear Materials Management and Safeguards System (NMMSS) data.
- PGDP historical site reports, including annual plant reports.
- PGDP reports describing facilities and production processes.
- Plant records.
- PGDP production records.
- PGDP analytical laboratory records.
- Correspondence between shippers and receivers.
- Historical DOE Draft and Historical Cultural Impacts of PGDP on the local area.
- The PGDP annual environmental survey reports on the site and other site investigation reports.

Few gaps were identified in shipping and receiving data. Where NMC&A data was unavailable, NMMSS data was used. Team members worked with representatives of other DOE sites with which PGDP interfaced via RU streams to verify shipping and receiving data and reconcile differences between sites where possible.

In addition to consulting the PGDP analytical laboratory records, the team found it necessary to glean analytical data from a wide variety of sources, including the PGDP historical quarterly reports. Correspondence between shippers and receivers also provided a record of comparisons of sets of analytical data, the first set developed by the site shipping RU and the second by the site receiving the material. In addition, analytical data has been compared and shared with other appropriate DOE sites.

For some areas that presented gaps in data that could not at present be filled by research, the project team developed estimates for quantities of RU and/or constituents. These estimates are based on extrapolations from actual data and represent (1) application of known data from similar material and/or circumstances or (2) application of known data from a specific time period over a longer or a shorter period of time. All such estimates and their bases are specifically identified in this report.

The approach used in searching for and collecting data useful to the project team's purpose was suitably comprehensive in terms of targeting the broad range of likely sources and locations of data. However, fundamental limitations currently remain that impact the team's capability to provide a comprehensively conclusive assessment at this time. Primary among these limitations was the short time available to search for, collect, integrate, and analyze information and data needed. Our team was limited to using annual role-up of U data reports due to time and document availability constraints; therefore no reconciliation of any discrepancies to monthly reporting document was preformed.

As a result of the brief but intensive search, the project team determined that a significant amount and quality of information still exists to address the scope and objectives established for this phase of the RU project. Further, results of this current effort have extended previous evaluations and have, in some instances, served to confirm earlier work. A significant amount of information

was identified, located, and collected as a result of this initial effort. More information has been confirmed or is thought to exist than has yet been collected. To address the scope and objectives defined for the project more fully will require significant additional work.

With respect to constituent analysis, some useful data was found, and more is needed and thought to exist. Even for the data found, there has been insufficient data and/or time to characterize the sample population with respect to the population it represents to the degree necessary to meaningfully employ statistical methods beyond simple descriptive statistics. Similarly, although PGDP established and applied quality assurance processes that the project team believes to have ensured generation of reliable data, statistical analysis of the data has not been performed and may not be possible to perform due to lack of complete data. Finally, in the short time available, priority was placed on obtaining and presenting referenced data useful to the project team's purpose. While estimates of uncertainty and confidence are currently unavailable, the data provides a basis for reasonable estimates and practical inference, and these have been made where possible.

Gaps are evident in the data in a number of areas:

- Where data exist for a specific constituent for a specific period, these data often represent a modest sampling of the stream.
- There are significant periods of time for which data for specific constituents have not been identified (or do not exist).
- Several Years of monthly Uranium Reports are missing and have not been discovered.
- Waste Management Records of storage and shipments are not readily available or have not been discovered.
- From the environmental clean up standpoint, most data are estimates and the site is still under site investigation to draw conclusions of past practices that would better identify release locations and sources.
- There are summary statements in some references (e.g., Smith and Bailey) that are purportedly based on data for which reference to the actual data is not included. These data should be confirmed.
- There are important streams for which constituent data probably exists, but have not yet been recovered.
- There are key potential areas of concentration for which constituent data is very limited or missing (e.g., feed plant ash, purge cascade traps, and feed cylinder heels)

## **6.5 Conclusions**

As discussed in Section 2.5 of this report, contamination control practices were lax at Paducah from the beginning of plant operation until the 1980's. Further, the use of PPE, particularly respiratory protection and company clothing was inconsistent. Often, the decision as to whether or not to wear a respirator was up to the employee. Based on these practices, it is clear that recycled uranium operations presented a significant challenge to the radiological protection measures actually used at the PGDP.

The recycled uranium processed at PGDP presented a potential for worker exposure greater than that attributable to uranium alone. (See Table 2.4-1) Feed plant processing of uranium oxides from Savannah River and Hanford tended to concentrate both Pu and Np in the ash by-product. Operations involving the feed plant ash represent the greatest potential or elevated exposure and release to the environment. Fluorinated Pu and Np from the feed plant would further concentrate in UF<sub>6</sub> cylinders and at the cascade feed points and lower stages. Worker exposures were

potentially elevated during cascade upgrade projects when equipment was moved that had TRU deposits. <sup>99</sup>Tc represents a reduced radiological hazard as compared to the uranium it was diluted within and does not appear to have concentrated in any unknown or unidentified process. It appears that the majority of Pu, Np and <sup>99</sup>Tc introduced to the Paducah Plant in recycled uranium was inadvertently released to the environment.

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KYL-28 Part 2, Analysis on Lots 122, 124, 126, 127-128; copy #10 of 16A (Hanford/Paducah), Dated: Dec 9, 1953

KYL-28 Part 3, Analysis on Lots 129-133; copy #10 of 17A (Hanford/Paducah), Dated: Dec 23, 1953

KYL-28 Part 4, Analysis on Lots 134, 135, 136, 137, 138; copy #10 of 16A (Hanford/Paducah), Dated: Jan 12, 1954

KYL-28 Part 5, Analysis on Lots 139-143; copy #10 of 17A (Hanford/Paducah), Dated: Feb. 10, 1954

KYL-28 Part 7, Analysis on Lots 150-154; copy #11 of 18A (Hanford/Paducah), Dated: Mar 19, 1954

KYL-28 Part 8, Analysis on Lots 155-159; copy #11 of 19A (Hanford/Paducah), Dated: Apr. 9, 1954

KYL-28 Part 10, Analysis on Lots 166-168, 171-172; copy #11 of 19A (Hanford/Paducah)

KYL-28 Part 15, Analysis on Lots 198, 200, 202, 204, 206; copy #10 of 18A (Hanford/Paducah), Dated: June 29, 1954

KYL-28 Part 16, Analysis on Lots 210, 213-214, 216; copy #10 of 19A (Hanford/Paducah), Dated: Oct. 12, 1954

KYL-28 Part 17, Analysis on Lots 219, 220, 222, 224; copy #10 of 18A (Hanford/Paducah), Dated: Oct 25, 1954

KYL-28 Part 19, Analysis on Lots 232, 234, 236, 238, 240; copy #10 of 18A (Hanford/Paducah), Dated: Dec. 6, 1954

KYL-62 Part 1, Analysis on Lots 4 – 13; copy #8 of 14A (Hanford/Paducah), Dated: Mar. 28, 1955

KYL-62 Part 2, Analysis on Lots 14- 18; copy #8 of 14A (Hanford/Paducah), Dated: Apr. 1, 1955

KYL-62 Part 3, Analysis on Lots 19 – 24; copy #8 of 14A (Hanford/Paducah), Dated: Apr. 11, 1955

KYL-62 Part 4, Analysis on Lots 41- 45; copy #8 of 14A (Hanford/Paducah), Dated: June 2, 1955

KYL-62 Part 5, Analysis on Lots 46 – 50; copy #8 of 14A (Hanford/Paducah), Dated: June 29, 1955

KYL-62 Part 6, Analysis on Lots 51 – 68; copy #8 of 14A (Hanford/Paducah), Dated: Aug. 12, 1955

KYL-62 Part 7, Analysis on Lots 69 – 95; copy #8 of 14A (Hanford/Paducah), Dated: Oct. 25, 1955

KYL-269 Part 1, Analysis on Lots 151-155, 157-159, 160-162, 163-165, 178-180, 193-195; copy #4 of 9A (Savannah River), Dated: May 10, 1962

KYL-269 Part 2, Analysis on Lots 208-210; copy #6 of 8A (Savannah River), Dated: Jun. 18, 1962

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KYL-166, Analysis on Lots UA-216, UA-220, UA-231 through UA-245; (Hanford/Paducah),  
Dated: Mar. 2, 1959

KYL-166, Analysis on Lots UA-256 through UA-265; (Hanford/Paducah), Dated: Apr. 30, 1959

KYL-166, Analysis on Lots UA-276 through 280, UA-291 through UA-295; Hanford/Paducah),  
Dated: Aug. 10, 1959

KYL-166 Part 11, Analysis on Lots 301-310; (Hanford/Paducah), Dated: Oct. 8, 1959

KY-L-411, Estimates of Transuranium Alpha Fed to Paducah Cascade, April 19, 1966

KYL-1239, Historical Impact of Reactor Tails on the Paducah Cascade, March 1994

**PGDP Site Team Approval Sheet**

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